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THE TRANSURANIUM PEOPLE

Imperial College Press



The authors (*left to right*): Dr. Albert Ghiorso, Dr. Darleane C. Hoffman, and Dr. Glenn T. Seaborg.

THE TRANSURANIUM PEOPLE

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THE TRANSURANIUM PEOPLE: THE INSIDE STORY

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Glenn Theodore Seaborg (1912–1999)

Glenn Theodore Seaborg, internationally renowned nuclear chemist, Nobel laureate in Chemistry, educator, science advisor to ten U.S. Presidents, humanitarian, and avid hiker, passed away at the family home in Lafayette, California, on Thursday, February 25, 1999, following a stroke suffered while attending the American Chemical Society meeting in Boston in August 1998.

Glenn was born in Ishpeming, Michigan, on April 19, 1912, of Swedish ancestry, a heritage of which he was very proud. When he was 10 years old the family moved to a small town near Los Angeles, because his mother wanted to expand her children's educational opportunities and horizons. He attended high school in the suburb of Watts and in 1929 entered the University of California at Los Angeles (UCLA), where he received his bachelor's degree in Chemistry and then stayed on for a fifth year to take graduate courses in Physics. In August 1934 he began graduate studies in Chemistry at the University of California at Berkeley (UCB) at a most propitious time following the invention of the cyclotron, in what he described as a "glamorous, exciting atmosphere." He completed his Ph.D. degree in May 1937, became the personal research assistant of Prof. G.N. Lewis, and joined the UCB faculty.

In early 1940, shortly after Edwin M. McMillan and Philip H. Abelson discovered neptunium (element 93), McMillan started looking for element 94 in bombardments of uranium with deuterons in the 60-Inch Cyclotron, but he was suddenly called away to wartime radar work at MIT before he could finish the project. Glenn contacted him and received his generous permission to continue this research,

and in February 1941 Glenn, together with his first graduate student, Arthur C. Wahl, and fellow instructor, Joseph W. Kennedy, produced and chemically separated plutonium.

Seaborg joined the wartime Manhattan Project in 1942 to work on the Plutonium Project, and when the decision was made that he should move to Chicago, he proposed to Helen Griggs, then E.O. Lawrence's secretary. They were married on June 6, 1942, and Seaborg often fondly called Helen "his greatest discovery of all." She and five of their six children survive him.

In 1944 and early 1945, the discovery of the new elements americium and curium was made possible by Glenn's recognition that they were part of an "actinide" series, and, thus, being trivalent in aqueous solution, could be chemically separated on that basis. He published his famous periodic table showing the "actinide series" in *Chemical & Engineering News* in 1945, the same year he returned (taking Albert Ghiorso with him) to become a professor of chemistry at UCB. Seaborg and his group discovered berkelium and californium in 1949–1950, and in 1951 he shared the Nobel Prize in Chemistry with McMillan for research on the transuranium elements. He was to be codiscoverer of five more elements: einsteinium (99) in 1952; fermium (100) in 1953; mendelevium (101) in 1955; nobelium (102) in 1958; and element 106 in 1974. The name for this last element, "seaborgium," was approved in 1997 by the International Union of Pure and Applied Chemistry, and Seaborg regarded this as an even greater honor than the Nobel Prize.

Seaborg served as Chancellor of UCB from 1958 until 1961, when he was chosen by President J.F. Kennedy to chair the U.S. Atomic Energy Commission, a position he held until 1971. He led negotiations resulting in approval of the limited nuclear test ban treaty in 1963 and led delegations to a multitude of countries to promote the peaceful uses of atomic energy. After his return to Berkeley in 1971, he resumed teaching duties, which he carried out until 1979, supervising the Ph.D. research of more than 65 students. He founded the Lawrence Hall of Science in 1982. He authored more than 500 scientific articles and numerous books. Because he had an acute

sense of history, he somehow also found the time to edit his valuable extensive and detailed journals, faithfully kept throughout his long career.

It should also be mentioned that Glenn loved sports and was a staunch supporter of the athletic program at Berkeley. Glenn and Helen loved to hike and laid out an interconnected network of 12-mile trails in the East Bay Hills above Berkeley which, when extended to the California–Nevada border, formed a link in the cross-country trek of the American Hiking Society.

In spite of his legendary accomplishments, Glenn Seaborg was basically a humble person who always had time for students, family members, and even nonscientists who wanted to visit with him. He prepared as carefully for lectures to freshman chemistry classes as for appearances before audiences of renowned scientists.

Our last formal collaborative effort with Glenn resulted in this book. Of course, in typical fashion, he completed his portions long ahead of time, but we worked long and hard to get all the pictures in and have him check the final texts before he left for the August 1998 American Chemical Society in Boston, where he was to receive one of his last accolades. He had been voted as one of the “Top 75 Distinguished Contributors to the Chemical Enterprise Over the Last 75 Years” by the readers of *Chemical & Engineering News*. He was presented with this award at a huge ceremony and reception on Sunday, August 23 — the evening before he suffered the stroke which ultimately led to his death.

We have lost a treasured colleague who, besides being a mentor, an advisor, and a unique resource, was above all a dear friend. We will sorely miss him, but he will live on through his prolific writings and in the cherished memories of the hosts of students, scientists, colleagues, politicians, and lay people whom he has influenced.

Albert Ghiorso

Darleane C. Hoffman

April 1999

Preface

Intimate Glimpses of the Authors' Early Lives

P.1. Darleane C. Hoffman

The story of how I came to be a “transuranium person” and one of the coauthors of this book is not straightforward and certainly not predictable. If you believe in astrology (I don’t), perhaps the only omen is that I was born on November 8, a birthday that I almost share with Marie Curie and Lise Meitner. These two famous women pioneers in nuclear science were both born on November 7, although Meitner’s birth date seems not to be entirely certain. Of more importance is that I was born into a family that prized education highly — and I recall my mother giving me little “intelligence” tests at a very early age!

My father, Carl Benjamin Christian, was born on October 7, 1898, to Bessie Ingelena and Albert Gustav Christian on a farm near Decorah, Iowa, a predominantly Norwegian community, and his parents were both of Norwegian ancestry. He had an older brother and two younger sisters. My mother, Emma Elvina, was born on March 2, 1900, to Mary Jane (Taylor) Clute and Eugene Clute on a small farm in Elk Creek Valley near the little town of Colesburg, Iowa. This was a largely German community, although her parents were of English and Dutch descent. She had an older sister, Nellie, born in 1897, and a younger sister, Minnie, born in 1903. The three girls were left orphans when their mother died of “consumption” in 1906. Although Nellie tried to help care for them, their father was unable to cope with the situation and found refuge in alcohol. My

mother, who was an extremely attractive child, was soon adopted in February 1907 by a German couple named Emma Minnie and Henry John Kuhlman, whose only daughter had died as a child. The other sisters were taken into foster homes just before being taken off to the orphanage, but were never formally adopted. The Kuhlman's decided to rename Emma Elvina "Elvina Emma" to avoid confusion with her new mother's name, but somehow in the process of recording it on the adoption papers, it was recorded as "Elverna Emma" — and that is how it stayed! Her life in her new home was a happy one — her new parents adored her and she, now being the only child, was in her own words "spoiled rotten." However, she sometimes found it difficult to fit into the German community, in which German was spoken often at home, at Church, and even at school.

My parents both graduated from Upper Iowa University: my mother with degrees from both the School of Music and the School of Oratory, and my father in Mathematics and Education. They met again later in Chicago, where my mother was working and my father was taking graduate work in education at the University of Chicago. They were married a couple of years later, in June 1925.



Fig. 1. Elverna E. and Carl B. Christian, 1925.

Photos of them at that time are shown in Fig. 1. That August they settled in their new home in the small northwest Iowa town of Terril, where my father at age 27 had been elected to the position of superintendent of the consolidated public school there. I was born at home on November 8, 1926, with my mother's oldest sister, now a registered nurse, and my grandmother Kuhlman in attendance to care for us. I became very close to my grandma, as in subsequent years she stayed with us on many occasions and often took care of me. I also sometimes visited my grandparents Kuhlman in the big city of Waterloo, Iowa, where she would take me to the movies on Saturday afternoon. In the evening we would often sing and they taught me little songs in German.

I was a frequent, extremely young attendee at a variety of school functions in Terril, including sports events, high school plays, musical events, and box socials. Often a couple of the unmarried women teachers "roomed" in our home and would baby-sit me in the evenings when my parents had to go out to "adults"-only functions. It was a special treat for me to be allowed to visit in their rooms and talk with them. My only sibling, my brother Sherril, was not born



Fig. 2. Darleane Christian at five years old with baby brother, Sherril, December 1931.

until September 28, 1931, when I was nearly five years old. A picture of me taken with him just after my fifth birthday is shown in Fig. 2.

I did not start first grade in the Terril Public Schools until the following year, when I was nearly six years old, because my father was a firm believer that *nobody* should start school until they were five years old, and since my birthday was not until November, I had not been allowed to start the previous year. Of course, I could already read and was ready to start, but he surely could not make an exception for his daughter! I remember my first and second grade teachers very well, as they had been among those who had stayed in our home. They were both excellent, "no-nonsense" teachers who kept track of three different levels of reading and arithmetic groups and still found time to help those who needed extra attention or to give me extra work to do to keep me from being bored. In those years our newspaper carried items such as "First Grade — *In a phonics test given this past week five children made no errors. They were Lillian Walton, Darleane Christian, Betty Blum, Wayne Glover and DeLane Anderson.*" (I still correspond with DeLane, who was my best friend.) And "Our honor roll for this period is: Darleane Christian, 55 points; Mildred Wray, 52 points; Ruth Rouse, 44 points. There were three A's this period. Darleane Christian and Mildred Wray had A in numbers and Mildred Wray had A in drawing and handwork. Dean Higley and Wayne Glover have been promoted to the A Division." Maybe if we still gave frequent public recognition for excelling in academic subjects as well as in sports, our present day students would have more regard for learning! Similarly our poetry was sometimes published. One of my second grade contributions which should have convinced everyone that I was not to be a poet was "*Spring is here at last, / And the winter is long past, / April is here with flowers, / To bring May's fair flowers. / Birds are singing in the trees, / And back have come the bees.*" A photo of my second grade class together with the first grade class is shown in Fig. 3. I am in the third row from the bottom — the second from the right.

During these years the U.S. was in the middle of the Great Depression. Consequently, my father was not paid during the summer



Fig. 3. First and second grade classes (1933–34) at Terril, Iowa Consolidated Public School.

months, so a couple summers we lived in a cottage at nearby Lake Okoboji, where our family and my father's brother and his family lived in a cottage on the lake and ran the nearby gasoline station. This was a wonderful time for me, as my cousins were the same ages as my brother and I. We played we were "Indians" in the woods and I learned early to swim in the lake and developed my lifelong love of swimming. We learned to play various card games, read — and on special occasions went to a nearby amusement park where we were not allowed to go on the roller coaster! Another summer, several years later when I was about nine, just our family ran a concession stand on the beach, selling snacks, cold drinks, popcorn, and renting out boats. That summer I helped at the stand, baby-sat my little brother, and on occasion rescued toys and even a few toddlers that had floated out to deep water, sometimes being rewarded by their parents with "tips." We often went out in the boat fishing with my parents. My main incentive for this was to help with the rowing and if I would sit quietly for awhile, which I hated, I was then allowed to dive and swim off the boat in the deep water. These were idyllic times, in spite of having very little money to spend. I remember having saved some of my "tip" money and

debating whether to buy a new bathing suit (which I badly needed!) or a large beach towel. I finally chose the beach towel, because it was less expensive, I knew I wouldn't grow out of it, and once in the water no one could see my bathing suit anyway!

We also visited my grandparents Christian on the farm near Decorah, where I found out about farm life — watching the cows follow the lead cow home from the pasture, helping my grandma “milk” them, churning butter, feeding the lambs milk from a bottle, taking lunch out to the men working in the fields at harvest time, sitting atop the huge draft horses, and lying on a blanket on the grass on summer evenings to watch the stars and listen to my father's two sisters tell me about the constellations. Sometimes at Christmas we would drive to visit them and my grandpa Christian would bring the horse-drawn sleigh to pick us up at the main road to take us through the deep snow down the hill to their home — one of my most treasured experiences. In the summer of 1933, I was allowed to accompany my parents and my aunts on the drive to Chicago, where we stayed for several days to attend the World's Fair. What an unbelievable and fabulous experience for me!

In 1934, after my second grade in school, we moved to Coon Rapids, a somewhat larger town (about 1500 people at that time) south of Des Moines, the capitol city of Iowa. Again, my father was school superintendent, but he also took on the job of coaching the girls' basketball team, since no one else could be found to do it. My mother would go to the games and run the popcorn popper and sell popcorn to make money for the sports program and new uniforms for the girls. My father was very successful as coach and soon everyone was attending the girls' games and leaving when the boys' teams came on to play! I played on the girls' junior high team, but it became obvious I would never be a basketball star — at barely five feet tall I simply couldn't compete with the nearly six-foot forwards that were the mainstays of girls' basketball at that time! However, I participated in a wide variety of musical activities, learning to play the piano, playing the saxophone in the school band, and singing in the chorus and small groups. In the summers my friends and I

bicycled, swam at the local pool, played croquet, played the piano and sang, and I read nearly every book in the local library. I attended third through ninth grades in Coon Rapids and had many close friends and was president of the freshman class. So it was a great shock to me when my father accepted a position as Superintendent of the Public Schools in West Union in northeast Iowa at the end of my freshman year in high school. I'm sure this was partly because both my parents had grown up in that area and we would be closer to my father's aging parents, who lived only 30 miles away and not far from my mother's parents in Waterloo. We moved there in the summer of 1941 and I began my sophomore year in high school.

Mathematics was my father's teaching specialty and was always a favorite subject in our home. We were entertained on long drives by doing the squares of the numbers up to 20 in our heads and calculating square roots with pencil and paper. (My brother later went on to major in Mathematics as an undergraduate, but switched to Physical Chemistry as a Ph.D. student at Iowa State University.) I took all the mathematics courses our schools offered. I even took advanced algebra, although that meant I had my father as a teacher, which I considered a very difficult situation as I was afraid the other students would think I was being favored! Not only that, I didn't even dare ask for help with my homework. (I took a trigonometry course one summer by correspondence, since it was not offered in our school.) I continued my interest in music and art, learning to play the flute and oboe in addition to the saxophone, and became a member of several choral and instrumental groups which won prizes at high school music competitions. I also took private art lessons. At my mother's insistence, I participated in dramatics and oratory groups and although this training was no doubt good for me, it was not my favorite extracurricular activity.

By February 1941, Glenn Seaborg had already become a "trans-uranium person" when he, together with his colleague Joseph Kennedy and his graduate student Arthur C. Wahl, made the first chemical separation and unequivocal identification of the new element plutonium. But I was still in high school and oblivious to all

those developments. However, I vividly remember the bombing of Pearl Harbor on December 7, 1941, which was announced while I was at a Sunday afternoon Community Chorus rehearsal of the *Messiah* at the local high school. The subsequent two years of high school were darkened by the pressures of a global conflict as we watched all the men teachers and many senior students go off to war while at home we coped with rationing of food, gasoline, and a variety of shortages, and waited for the lists of war casualties. I corresponded with several young men throughout the war — fortunately, they all came home safely. My father tried desperately to enlist but was repeatedly turned down because of his age and position.

I graduated from high school in 1944. A young man and I were coaledictorians with the highest grade averages ever recorded in the high school there. I decided to enter Iowa State University at Ames, Iowa, but had difficulty trying to decide whether to major in Applied Art or Mathematics, but finally settled on Applied Art. In those days Applied Art was in the College of Home Economics, and fortunately for me, I was required to take Home Economics Chemistry. (I had never studied Chemistry as it was not offered in our high school, but I did take Physics from a substitute teacher and perhaps because of that found it less than exciting.) The beginning Home Economics Chemistry at Iowa State was taught by Prof. Nellie Naylor, and largely due to her outstanding teaching I found myself more interested in Chemistry than anything I had ever studied. She had a way of making it all seem so beautifully logical as well as relevant to a host of everyday problems. Consequently, I decided by the second quarter that I would switch my major to Chemistry. This somewhat unconventional choice caused my Applied Art Counselor to ask me, "Do you really think chemistry is a suitable profession for a woman?"! I replied that I was quite sure it was. After all my excellent Freshman Chemistry teacher was a woman. However, both these women were what we used to call "spinsters," and I vowed that I would not necessarily emulate them in this respect but would maintain other interests and continue to date a variety of young



Fig. 4. Darleane Christian, photo for Iowa State College Yearbook, 1945.

men and might even consider marriage as well. So in Spring quarter 1945, I became a Chemistry major in the College of Science. (See my class photo taken in Spring 1945 in Fig. 4.) From that time on I was usually the only woman in most of my classes, but this bothered me not at all, nor did it seem to bother the young men in my classes. After all, during the war years young men were in relatively short supply, although at Iowa State there were a number of officer training programs and the veterans began returning to school on the "GI Bill of Rights" at the end of the war.

By 1945, Seaborg had been leading the effort at the Metallurgical Laboratory in Chicago for several years to develop a process to separate plutonium from the large amounts of fission products and the uranium in which it had been produced by neutron irradiation. The process had been tested in a pilot plant at Oak Ridge and the large processing plants at Hanford Washington had gone into operation in December 1944. Albert Ghiorso joined the "Met" Lab in 1942.

During my first years at Iowa State (1944, 1945), we would often hear all kinds of wild rumors about what might be going on at "Little Ankeny." This was an installation of some rather drab, temporary-looking buildings on the edge of the campus from which

some rather spectacular flashes of light were seen to originate from time to time that illuminated the night sky. It was not until much later that I learned of the "project" at Iowa State College under the leadership of Frank H. Spedding and Harley A. Wilhelm to solve the problem of reduction of uranium to very pure uranium metal and deduced it had something to do with that (Spedding had come to Ames after receiving his Ph.D. at Berkeley under the direction of G.N. Lewis). For a time, Spedding was also director of the Chemistry Division of the Metallurgical Laboratory at Chicago. He was Director of the Institute for Atomic Research at Ames when I first joined that institute as an undergraduate research assistant in the summer of 1947, after my junior year at Iowa State. During Spring quarter of that year two openings for undergraduates in the Atomic Research Institute were posted and announced in our Chemistry classes. I especially remember this being announced in my advanced inorganic lecture class by Prof. John Wilkinson, a very hard taskmaster indeed, but one who encouraged me to apply, probably because I was one of the few students in his class who could quickly recite all the known acids of sulfur and phosphorus! Although I was somewhat pessimistic about my chances, I did apply because I was increasingly tired of working at odd jobs in the dormitory dining room, the Botany Department, and grading papers in the History Department, and figured I had nothing to lose! (Although I had a tuition scholarship, I still had to help with the expenses of my room and board.) To my amazement, I was called to an interview with Dr. Don Martin, Jr., Professor of Inorganic and Nuclear Chemistry, who had been at Los Alamos during the war. (Rumor was that he had ingested some quantity of separated polonium, although I never asked him if this was true.) He asked if I would be willing to help make and test the Geiger counters that he was building to be used to assay samples for radioactivity. I was thrilled with the idea and recalled the biography of Marie Curie I had read in eighth grade and how fascinated I had been with her studies of radioactivity and her painstaking isolation of the new elements radium and polonium and her use of radium in medicine. When I was offered the position and

found that as a full time employee in the summer I would, in addition to being able to do research in the Institute, earn \$170 per month for something I would have been happy to do for free, I was doubly thrilled. (This also released me from a very dull summer job as a bank teller in my home town for only \$85 per month, a salary I previously had thought was quite good as before that I had been a waitress in the local "best" restaurant in town at \$7 per week!) I still did not know about the discoveries of neptunium and plutonium, the first transuranium elements, but another plus of working at the Institute was that they obtained a Q-clearance for me. After that, I became privy to some of the new information that was coming out. One of the odd quirks that I encountered was that on my classified notebook I had to put three initials and since I had no middle name I chose the letter "X" and told them it stood for Xanthasia, which seemed to satisfy the system. I think it was with this position at the Institute that I really started down the inevitable path that led me to become a "transuranium person." I continued to work at the Institute part-time during my senior year, splitting mica for windows for the Geiger counters, annealing the copper electrodes, etc., and making measurements of various radioactivities. I also learned micro ion exchange resin column separations for rare earth elements (a research interest of Spedding, who had developed very large-scale column separations for the rare earths), and reduction and separation of certain lanthanides with sodium amalgam. Knowledge of these techniques later became very useful to me in the transuranium field.

It soon became clear to me that I wanted to continue to do research in nuclear and radiochemistry, and Prof. Martin suggested that he would be happy to recommend me for graduate school at his alma mater, the California Institute of Technology, but going to California was too big a step for me to take at that time. Furthermore, by then a new 68-MeV synchrotron was being completed at Ames that opened many exciting research possibilities, and I elected to stay at Iowa State and continue to do research with Martin. During my first year in graduate school I met my husband-to-be, Marvin Hoffman, who had just come to Ames from the Cyclotron

group at the University of Chicago, where he had been working for a year after returning from the Navy. He became a graduate student in nuclear physics of L. Jackson Laslett, then a Professor of Physics at Ames, who had been a student of E.O. Lawrence at Berkeley. Yes, indeed, it is a small world!

My father, although in accord with my decision to go to graduate school, suggested that I might want to get a teaching certificate in order to make sure I could get a job when I finished. This I absolutely refused to do, saying that the last thing in the world I ever wanted to do was teach!! Many times since I have thought how happy he would be to see that finally I recanted and eventually became a Professor of Chemistry at Berkeley, albeit via a most circuitous route. However, my decision to stay at Iowa State turned out to be a good one, as most unexpectedly my father died of a heart attack only two years later, at age 52. I was called at 4 a.m. by my mother, who told me the awful news and that the high school coach would drive to Ames that afternoon to pick up me and my brother, who was by then an undergraduate at Iowa State, and drive us home. I remember going in to ask if I could be excused from my classes and make up a midterm exam I was to have the next day in Quantum Chemistry when I returned. Instead, much to my dismay, my professor in Quantum Chemistry insisted on giving me the exam right then! Needless to say, I had a hard time concentrating on the test and hadn't really studied for it yet. I think he actually believed he was doing me a favor, and I managed to get a B in the test, but I never forgave him for his insensitivity. I went home to northeast Iowa to help my mother with funeral arrangements — she didn't drive, nor had she ever even written a check! My father's services were held in the high school gymnasium and some 1200 people attended. Although we were most appreciative of this indication of the community's respect for him, it was an ordeal I shall never forget as I strove to maintain my composure and never shed a tear. I later wrote a theme in a creative writing course vilifying all funeral services. I then became more or less responsible for my mother and

my brother, who continued his studies as an undergraduate in Mathematics at Iowa State.

My mother and I had to clear out the house quickly, as it belonged to the school district and they needed it for the next superintendent, and so we kept what little we could and had an auction to get rid of the rest. She then came to live with me in Ames for awhile and soon after became house mother at one of the fraternities there. She was later nominated by her fraternity "boys" and named Ames "Mother of the Year." I continued my graduate studies (see Fig. 5) on photonuclear-induced Szilard–Chalmers reactions at the Synchrotron. My friendship with Marvin Hoffman was furthered by the fact that he was working at the Synchrotron and able to run it for me in the evenings so I could get irradiations almost anytime I needed them! I used a variety of complexes of cobalt and platinum which I synthesized to enable me to obtain very high specific activities and discover several new isotopes of cobalt, platinum, and iridium. For me, the discovery of new isotopes which nobody had ever seen before was an exhilarating experience, and to this day I still find the discovery of new isotopes — or, even better, new chemical elements — the most exciting part of nuclear chemistry research, although it is getting to be more and more difficult to do!



Fig. 5. Darleane Christian, Spring 1950 photo at Ames Laboratory, with remote apparatus for pipetting radioactive solutions.

I finished my Ph.D. in December 1951, in just over three years, and Marvin and I were married on December 26, 1951, in Waterloo, Iowa, the home of my grandmother Kuhlman. In January 1952, I left Marvin behind to finish his Ph.D. in Physics and I went to Oak Ridge to begin my new position there on the Aircraft Nuclear Propulsion Project, which involved uranium, but still not transuranium isotopes. I found out later that Prof. Laslett had told Marvin it was a terrible mistake for us to get married and that the marriage would never last under such unconventional circumstances! Some 20 years later, I had the pleasure of entertaining him in our home in Los Alamos, and again in Berkeley in the 1980s, which I think convinced him otherwise. (In spite of that erroneous prediction, we both thought he was one of the most intelligent men we ever met!) Marvin finished his Ph.D. near the end of 1952, and he decided to take a position in the Test Division at the Los Alamos Scientific Laboratory, where he had worked as a summer graduate student assistant in 1950. He was told that I would be offered a position in the Radiochemistry Group of the Test Division, so I quit my job at Oak Ridge and we went to Los Alamos after Christmas in 1952. Although I spent only a year at Oak Ridge, I made many close friends there. Once in Los Alamos, I immediately started calling the personnel department to ask about my job in the Radiochemistry Group of the Test Division and they told me, "There must be some misunderstanding, we don't hire women in that Division." Having never before run into such discrimination, I was totally taken aback and asked them to please try to circulate my application and find out where my job was supposed to be, but to no avail. Finally, in January 1953, Marvin and I went to a cocktail party for new hires and their spouses hosted by Director Norris Bradbury and I met Dr. Roderick Spence, group leader of the Radiochemistry Group. We talked and I told him my story and he said, "Where have you been — I've been looking for you. We need you for plutonium chemistry." Mike, the first thermonuclear test, had been fired in November 1952, in Eniwetok, and they were busy analyzing its unexpected and exciting results and badly needed more radiochemists, especially someone to

devise new separation procedures for plutonium. So he hired me the next week, greatly relieved to know that I had a clearance and could start work immediately. Nuclear chemist Charles I. Browne, Ph.D., 1952, University of California, Berkeley, had joined the group as a military staff member in September 1952 and H. Louise Smith, M.S., 1952, University of Kansas, Lawrence, Kansas, had been in the group only about a week before the Mike test samples began to arrive! Rod Spence himself had been in the Pacific when the test was conducted and was there for the collection of air samples of the debris on filter papers held in special samplers attached to the wing tanks of airplanes which flew through the resulting cloud of debris. Little did we know that it would be mid-March 1953 before I would actually be allowed to get started. It seems my clearance was "lost" between Oak Ridge and Los Alamos. Since I originally got my clearance at Ames, it went back to the originating office rather than to the office which handled the Oak Ridge clearances. So they couldn't find it for three months — finally, after calls from everyone to Personnel, they asked the FBI to start a new clearance and they found it in the Chicago office in about three days. So that is how I missed being a discoverer of einsteinium ($Z=99$) and fermium ($Z=100$), which were identified in the debris from the Mike test while I was sitting in a small apartment in Los Alamos raging at the system. I will never again trust personnel offices, not just for saying "we don't hire women in that Division," which was untrue, but for their general insensitivity, incompetence, and bias — qualities which were not generally shared by the male scientists with whom I have worked!

Anyway, I finally did join the Radiochemistry Group at Los Alamos, on March 13, 1953. My first project was to find better and faster methods for the separation and analysis of plutonium in debris recovered from tests in the Pacific and later from above ground tests at the Nevada Test Site. Rod Spence was a wonderful mentor for me and taught me a great deal. I admired him greatly not only for his scientific ability but because he was one of the fairest and least egocentric individuals I have ever known. At last I had become a genuine "transuranium" person. The whole story of the

discovery of elements 99 and 100 in the debris from the first thermonuclear device tested in November 1952, is given later (Chapter 6) in the book. The story of how I eventually became a “transplutonium” and even a “transactinide” person will unfold as the book progresses.

I should not fail to mention that during the days in Los Alamos Marvin and I produced two children. Our daughter, Maureane, was born on Easter Sunday, 1957, and our son, Daryl, was born on September 2, 1959. Los Alamos was a wonderful place to raise children. In both cases, I was able to continue my work until a day or two before each of my children was born and then quickly go back to work afterwards. I was privileged to have a wonderful woman to take care of them during the day. Then in 1964 my mother came to live near us after her mother passed away and she was instrumental in making it possible for me not only to pursue my career, but to travel as necessary. However, both Marvin and I spent as much time with our children as possible. A 1974 photo of one of our leisure activities is shown in Fig. 6. Although Marvin was not musical, he was a great listener and critic! Both of our children went to the Los Alamos schools and graduated from high school there.



Fig. 6. Darleane, Marvin, Daryl, and Maureane Hoffman around the piano in their home in Pajarito Acres, Los Alamos, NM, 1974.

Maureane received her B.S. from New Mexico State University in 1976 and an M.D. in pathology and a Ph.D. in Toxicology from the University of Iowa in 1981. She is now a tenured professor in the medical school at Duke University, in Durham, North Carolina. Daryl received his B.S. in 1981 from the University of California, Los Angeles, an M.D. from the University of New Mexico in 1984, completed a six-year residency in plastic surgery at Stanford University in 1990, and is now in private practice in the Palo Alto, California, area.

Two singular events which proved to be career-shaping should also be pointed out. The first is that in 1964 I was awarded an NSF Senior Postdoctoral Fellowship and Marvin was awarded a Fulbright Fellowship to Norway, so we took our two children and my mother and went to live in Oslo for a year. I found this a personally very liberating experience, as in Norway women were treated equally, but they were also expected to take their share of the responsibility as well. It was quite safe to go places by oneself and also not unusual for women to go out alone, while at that time in the U.S. women usually didn't dine out at restaurants alone or go on business trips with male colleagues. I performed research on short-lived fission products at the reactor at the Institute for Atomic Research at Kjeller, near Oslo, and learned new rapid separation techniques. We made many long-time friends there and were able to visit the farm at Havaas on Hardangerfjord, where my grandmother Christian's parents came from, and the area in Gubrandsdalen, where my grandfather's parents came from. Our daughter attended second grade in the neighborhood school and became fluent in Norwegian. We returned to Los Alamos a year later — all feeling greatly enriched by the experience.

Much later, I was awarded a Guggenheim Fellowship for the year 1978–79 for the study of the mechanisms of nuclear fission, and I spent this sabbatical year with Glenn Seaborg's group at Berkeley and Marvin had a research position at SRI International in Menlo Park, California. I had already become well acquainted with Glenn Seaborg after my successful search for ^{244}Pu in nature in 1971, and

my appointment in 1974 to the first IUPAC/IUPAP *ad hoc* committee to consider claims of priority of discovery of elements 104 and 105. I also worked closely with him (mostly by telephone, since I was still in Los Alamos) in the final honing and careful wording of the 1976 Science "Criteria" article [9-3]. During this time I had the opportunity to work with Al Ghiorso and Diana Lee, both so important to my future career. During this time also, Al Ghiorso designed and we built and tested the Merry-Go-Around (MGA) rotating wheel system (Fig. 7) which we used to study the spontaneous fission (SF) properties of the isotopes, fermium-246 and -248, which had half-lives of only a second and 36 seconds, respectively. The MG was later upgraded to study α -decay and used in a special "mother-daughter" stepping mode devised by Ken Gregorich for the 1993 seaborgium confirmation studies (Chapter 10). During the year 1978-79 I also became involved in some of the searches for superheavy elements (SHE) and attended the brown bag lunches of the "Super Heavy Element Isotope Kjemikers" (or SHEIKS) held in Seaborg's office every Wednesday noon. When I left Berkeley in July 1979 to return to Los Alamos to become Division Leader of the Chemistry-Nuclear Chemistry Division, the SHEIKS group had a wonderful party for



Fig. 7. Darleane Hoffman and Diana Lee with the MGA at the LBL 88-Inch Cyclotron, 1979.



Fig. 8. Darleane Hoffman cutting a cake at her going-away party, July 1979. Matti Nurmi is at the right.

me with a cake on which was written, "It was SHEer pleasure knowing you, Darleane". My picture with this lovely cake is shown in Fig. 8. I think that this pun was probably the brainchild of either Matti Nurmi or Al Ghiorso, who were our resident experts at devising clever acronyms, puns, etc.

I reluctantly cut my sabbatical year a few months short in order to return to Los Alamos in the late summer of 1979 to become the Division Leader of the Chemistry–Nuclear Chemistry Division — the first woman to fill such a position at LASL. This was a great challenge and honor for me and although I hated to leave Berkeley, I was eager to take up my new position. I had many ideas about things I wished to implement in chemistry and nuclear chemistry — not just in heavy elements, although certainly I had been seriously inoculated with that virus. When I could, I took time off from my administrative duties at Los Alamos to go to Berkeley (and even GSI) to participate in fission studies of the fermium isotopes and searches for SHE. It was a very busy time for me, but also very productive.

My Los Alamos colleagues and others nominated me for the 1983 ACS Award for Nuclear Chemistry "for her contributions to the understanding of the forces that govern nuclear behavior through

studies of the fission process and of the production and characterization of heavy elements, both man-made and in nature." And, indeed, I was chosen for this award, the first woman selected for an ACS scientific award, other than the Garvan Medal, which is specifically designated for a woman chemist. (I was also very pleased to receive the Garvan Medal in 1991, but I was especially pleased that I was first honored by my colleagues for my ability as a nuclear chemist rather than because I was a "woman" chemist, prestigious though that award is.) As fate would have it, much to my pleasure, I learned that Glenn T. Seaborg was to present this award on March 21, 1983, in Seattle, Washington, during the National ACS meeting there. The picture of us taken on the occasion of this Banquet and Award ceremony is shown in Fig. 9. A color version of this hung in Glenn's office at LBL for several years until, much to my regret, he replaced me with a picture of himself and the movie star Ann-Margret!



Fig. 9. Glenn T. Seaborg presenting the ACS Award in Nuclear Chemistry to Darleane Hoffman, March 21, 1983, at the ACS National Meeting in Seattle, Washington.

Probably largely as a consequence of my 1978–79 sabbatical year at Berkeley and my subsequent close association with the heavy element group, in 1984 I was invited to return as a Professor in the Department of Chemistry of the University of California, Berkeley,

with an appointment at LBL as Faculty Senior Scientist and Leader of the Heavy Element Nuclear and Radiochemistry Group. It was in some ways a difficult decision for me, as I was very devoted to my Division at Los Alamos, but I felt they were in good shape and that it was time for me to help educate the next generation of students in nuclear and radiochemistry. And I also wanted to pursue my interest in the chemical and nuclear properties of the heaviest elements and maybe even help search for new elements. Berkeley was the ideal place for that. So I accepted and then Marvin and I began the difficult process of moving away from Los Alamos after more than 31 years there. So I came to Berkeley in August 1984 and I started my next career as Professor of Nuclear Chemistry. I continued my close association with those giants and pioneers of nuclear science, Glenn T. Seaborg and Albert Ghiorso, with whom I am now privileged to coauthor this book.

P.2. Albert Ghiorso

I was born on July 15, 1915, in Vallejo, California, as the fifth of the seven children that my mother would have. Two died in infancy, leaving our family with three girls and two boys to grow up together in Alameda just across the bay from San Francisco. My father, John, had emigrated from Genoa, Italy, with his family when he was two years old. The family, my grandmother and grandfather with four sons and two daughters, settled on a very small farm in the hills above St. Helena and there they eked out a modest living. Thirty years later I would spend most of my summer vacations at this ranch. My father left the ranch when he grew up and became a jack-of-all-trades, making his living at various times as a taxi-driver, riveter, welder, cook, handyman, etc. Although he never attended school beyond the fourth grade, he knew a lot of lore and respected education. Like most working men of that time, he was a strong union supporter and a political radical and wanted his children to amount to something in their lives. In particular, he wanted me to become a lawyer — an *honest* lawyer, he emphasized!

But that was not in my plans. Although I was good at such things as history and the other subjects that a good student can excel at, I also had a mechanical aptitude that showed up very early. I remember at the age of about five playing with shingles discarded by carpenters who were building houses nearby. I became adept at structures and learned how to make things. One incident that stands out in my mind occurred on my grandfather's ranch. An old automobile had developed a flat tire which had to be pumped up manually. The pumping hose would not stay in place and I was told to hold it there with my hand. I noticed that when the pumping commenced, the hose became very warm, and this aroused my curiosity. I asked my uncles why this happened and they did not know. I soon figured out that it was because the act of compressing the air had heated it. When I explained it to the adults they marveled that a mere child could know these things.

I never became interested in the technical side of radio as a child, although my father tinkered with the new-fangled invention and gradually acquired a lot of equipment as a hobby. Some years later this gear was to have a powerful influence on my career. Instead of radio, I loved making gliders and rubber-powered model airplanes that flew and I gradually learned the rudiments of design by trial and error. For a long time I had set my sights on becoming an aeronautical engineer, undoubtedly influenced by the close proximity of the Oakland Airport. I became a member of the Aviation Club at Alameda High School and was vice-president of the eight-member club in 1932 (Fig. 1). Our house in Alameda was at the east end of that island, directly across San Leandro Bay from the airport. I used to bicycle the two miles to the airport regularly to examine the planes at close range. I remember quite vividly being present with hundreds of thousands of others when Charles A. Lindberg landed there on his barnstorming tour of the country after his historic flight across the Atlantic in May 1927.

My mother was born in Watsonville, California, of pioneering stock that stemmed from the Spanish Land Grant times. She was a tender, pious woman, and made sure that I was baptized in the



Fig. 1. Photo of Ghiorso cropped from a picture of the Aviation Club included in the 1932 Alameda High School yearbook, *Acorn*.

Catholic Church despite the fact that my father was not only openly antagonistic to the Church but was also an avowed atheist. I did what my mother wanted me to do and attended Sunday school dutifully and absorbed the dogmas as a willing believer. The only books as a child at home that I had around me were religious tracts and I read all of them voraciously. By the time that I started to attend school and had access to libraries I was quite happily indoctrinated. That was not to be challenged until I was 13 years old when, in a high school history class, I realized that most of the world was not Christian and had beliefs and superstitions that were quite different than those that I had been ingrained with.

I did well in school and always assumed that I would get into some kind of academic profession. However, when I graduated from Alameda High School in 1932 the Great Depression had gripped everything very tightly and our family did not have even the modest sum (\$26 incidental fee!) that it would take for me to enter the

University of California. My father was out of work most of the time and had to resort to bootlegging liquor to keep going, a stratagem used by many people during Prohibition. Meanwhile, my “Big” sister, Genevieve, had applied for a small scholarship for me to go to Cal and this made the difference. Now I could commute to the University by virtue of the excellent public transit system (light rail!) and live at home. But which one of its colleges should I enroll in?

I had a whole summer to decide. Since there was not even a summer job available for me, I had nothing to do except read and think. Among the books that I got from the local library was one called *Letters from a Radio Engineer to His Son*, by John Mills, published in 1922. Mills, a Western Electric Company engineer, wrote this fascinating book of 24 letters to explain to his son in a very straightforward, detailed way how radio worked, and he expanded his tutorial by suggesting simple experiments that could be undertaken to demonstrate the new science. I decided to try my hand at them, since I had the legacy of the gear that my father had played with in the 20s.

The book had a profound effect on me by uncovering a whole new world that I would marvel at and I proceeded to construct the experiments one after the other. Thus encouraged, I enlarged my sphere of knowledge and skills even further by poring over the hobby magazines to make small pieces of simple equipment. I remember making a supersensitive carbon microphone, a thin pencil carbon resting vertically and loosely in indentations in two blocks of carbon that could detect a fly walking on the sounding board (cigar box) that supported it. The experiments were simple but elegant, in that they instructed me in some of the basic principles of radio. I concentrated all of my efforts day after day on learning all about this new science from magazines as well as by doing the experiments; by the end of the summer I knew that I wanted to be a radio engineer. However, there was no such category in the university’s curriculum at that time and I had to settle for electrical engineering. Sixty years later I told this story of how my career was started to an ORNL audience and it struck a responsive chord with my good friend Dave

O'Kelley, who knew that book very well, also as a youth. He went to some trouble to find a copy of the book which had been out of print for decades and sent it to me. I treasure it as the beginning of a new life for me.

My career at UC was not spectacular; it seemed that all of the engineering students were excellent and this raised the class averages such that a high school student who was top dog in some small school did not necessarily rise to the top in a class of a thousand others. Still the courses were usually interesting, because I encountered so much new information. I managed to perform as an above average student for the four years and learned how to think scientifically. I particularly enjoyed freshman chemistry, especially the lectures by the inimitable Joel Hildebrand. My senior year was really exciting, though, because now I could specialize in courses that would affect me more directly. I remember auditing a course given by Prof. Lester Reukema in which he devoted the better part of a semester to the subject of negative feedback. This brand-new invention by Black at the Bell Telephone Laboratories in 1936 was one of the great discoveries in electronics; it was to have a profound influence on the field of amplification and spread to all other fields as well. I attended the Physics Department Journal Club occasionally and heard about some of the new discoveries that were being made by Ernest O. Lawrence's invention of the cyclotron.

I graduated in May 1937 ready to go out into the world and become a great radio engineer, but there were no jobs available as far as I could tell. This was about the low point of the Great Depression. I survived because I still lived at home in Alameda. I got small jobs making amateur radio equipment to order for "hams" who wanted special receivers or transmitters. I had developed a modest reputation in this regard because of my illegal operation of a "bootleg" radio transmitter on the five-meter band. Most unusual for the time, I had built a crystal-controlled transmitter and a superheterodyne receiver for this band to work with a rotating beam antenna and these had proved their superiority over the conventional equipment employed by others.

In particular, this became clear when my brother, Gilbert, and I contacted an amateur station in Ohio for a few minutes one day when the skip distance happened to be just right to bridge the signals across the 25000 miles that separated Ohio from California by bouncing them off the Kennelly–Heaviside layer. It was the first time this had ever been done on the five-meter band and was mind-boggling to everyone since it was well known that the radiations in this band only traveled in straight lines. This was my first venture into the field of discovery, but I never got credit for this exploit since the station was operating without a license. We had borrowed the call letters of a friend.

However, my reputation was now made. Since none of the equipment that I used was available commercially at that time I was asked by a wealthy executive, Larry Barton, of the Clorox Chemical Company, who was a prominent radio amateur, W6OCH, to build him a fancy receiver. He was willing to pay whatever it took and I immediately set about designing and building his receiver.

With this entrée into the world of high class radio equipment I came to the attention of D. Reginald Tibbetts, W6ITH, who was famous as the biggest powerhouse in the world among all radio hams. Having nothing better to do, I went to work for Tibbetts to gain experience. In Berkeley he ran a small business, Communications Supply Co., that catered to special needs. He had pioneered two-way communication setups for the building of the SF-Oakland Bay Bridge, and with his ability now well established he had begun to supply portable radio equipment for emergency services. All of this gear was highly specialized, so it had to be built to order. My job was not only to build but also to design and I had lots of opportunities to create new things to fit the needs that arose. One interesting project was to design and build transceivers for use in the construction of the Shasta Dam in California. The objective was to communicate between the Head Tower and the enormous “buckets” that carried the concrete that was poured into the giant forms. The work was done around the clock and reliability was the keynote. The equipment worked fine but developed problems in the field because the units were housed in

small black metal cabinets. In the hot sun that beat down into the canyon the temperature rose high enough to melt the capacitors. I spent several weeks in the area one summer until I had brought everything under control.

When there was no outside job that needed my talents, I worked on Reg's "ham" transmitter and receiver facilities. These were very elaborate, of course, so it was fun for me to work on them. By 1940 he had moved out to Moraga into a small valley which was ideal for setting up giant rhombic antennae aimed to all points of the compass. This enabled him to transmit to and receive from stations around the world. One of the problems that I was confronted with was how to measure accurately the frequencies of the stations that were received. I conceived of a simple way to accomplish this by measuring the beat frequencies between the received signal and locally generated crystal-controlled signals (which came in exact 10 kHz intervals). I measured the beat frequency with a local precision audio oscillator. This device was very successful and in due time we were asked to submit an article for *Electronics Magazine*. Tibbetts and I were supposed to be coauthors even though the idea and its reduction to practice were mine, but when the article appeared my name was missing. Reg blamed the magazine for the oversight but I doubt that the fault was theirs. He gave me the \$30 that was the going rate for articles at that time, probably out of guilt that he had left my name off the article. That was my very first publication and a good one at that.

Although I was not paid very well, I enjoyed the job and gained invaluable experience from working on so many different projects. One of the most valuable to me turned out to be the engineering and installation of an intercommunication system at the Radiation Laboratory on the Berkeley campus of the University of California. This was set up in 1940–1941 to connect the secretarial desks and I soon made the acquaintance of the two most important people in the Lab as far as I was concerned. One was Helen Griggs, who was Lawrence's secretary, and the other was Wilma Belt, who was Donald Cooksey's secretary. Cooksey was Lawrence's deputy and ran the

logistics side of the Lab in terms of personnel and special materials that had to be ordered or made in the Rad Lab shops. I got to know both of these women fairly well and before long discovered that Wilma and I had many common interests.

In 1941 Tibbetts was asked by the Rad Lab to produce what turned out to be the world's first commercial Geiger-Mueller counter circuit. It was to count particles with a scale of eight and a mechanical register and to have a regulated high voltage power supply for the G-M counter. It seemed that for some reason I was not told that it would be necessary to build hundreds of these devices for Prof. Glenn T. Seaborg's group. It was clear to me that I would have to set up some sort of assembly line to produce the required number in the short time allowed for delivery. The circuit diagram had been published in the *Review of Scientific Instruments* and a working copy of it had been made by one of Seaborg's chemists, Dr. Joseph Kennedy, as a model for us to produce on a mass scale. I remember with horror seeing the model chassis. The circuit was all crammed together in a space that was quite limited, about 12" \times 17" \times 8". None of the resistor or capacitor leads had been trimmed and everything was jammed together with no room for anything else. However, it did work and I soon decided to re-engineer it so that I could build the units with some assurance that they would all work. This turned out to be quite a job, but a very interesting one since I had never done anything like this before. I was to leave Cyclotron Specialties Co. in about a year and in that time I built some 300 of these units for the Manhattan District Atomic Energy Project. I often visited the Rad Lab for various reasons and I became fairly well acquainted with Seaborg's scientists, in particular Spofford English and Gerhart Friedlander, who became my mentors. Seaborg, himself, I knew only casually.

Wilma was another matter. I came to know her very well and soon we began dating. She was particularly interested in "hot jazz," something that I knew nothing about. We listened to records and went to the Dawn Club in San Francisco to hear Lou Waters and His Band. I, too, soon became an aficionado of this old/new music. We

also were both interested in the outdoors and we had planned a Sunday (December 7, 1941) for a trip to Yosemite Valley to see what it was like in the winter. We left early in the morning to make the 200-mile trip in my old Chevrolet coupe. In four or five hours we were in the Valley and what a sight that was. There was snow everywhere, no traffic, and no people. It was truly a winter wonderland. Wilma had prepared food and we spent the day exploring the park, never once turning on the radio. We were not the least interested in what the outside world was doing anyway! We were deeply in love. We already knew that WW2 was going badly in Europe and it seemed far, far away.

The end of the day all too soon made it necessary for us to wander back to civilization. An hour's drive took us down into the Central Valley, where finally we turned on the car radio for the first time. What a cacophony there was! For fully two hours all we heard was the fact that all sorts of troops were being called up. Why this was being done was not being discussed at this time. Something fearful must have happened early that morning. Finally, we heard the news. A surprise attack had been made by many planes from Japanese aircraft carriers on a good part of the Pacific Fleet quietly at anchor in Pearl Harbor. They had succeeded in essentially knocking out a large part of the big guns of the American Fleet in the Pacific! Panic was striking the West Coast! Was invasion near? The news was terrifying to us. Obviously, our country was now completely engulfed by WW2.

By the time that we reached Berkeley we had discussed over and over what might happen to us. To forestall the worst scenario we decided to get married as soon as possible. We would worry later about the inevitable draft that was bound to sweep me into the armed forces. For the time being we expected that I would probably be deferred at least temporarily by the work that I was doing for the Radiation Laboratory.

We were married the following month and enjoyed several months of bliss. But the war became more and more grim as the Japanese continued to win everything in sight in the Pacific. Everyone could

see that it was going to be a long hard battle for the U.S. to regain the lost territory. Soon I felt that my chances of avoiding the draft were becoming vanishingly small and I decided that rather than be drafted into the regular army, where my skills would be completely lost, I would be better off trying to obtain an officer's commission in the Navy. There I had a chance to be more useful, I assumed. Accordingly I applied for a Lieutenant, JG, commission, since I had heard that all it took was a college degree and recommendations from a couple of prominent people. Wilma suggested that I use Seaborg as a reference as he was a university professor.

I agreed that this was a good idea even though I did not know Seaborg very well, and I wrote a letter to him, now in Chicago, where he was setting up his group. He sent me the recommendation letter that I needed but he also made me an offer to join his group. He said that he could not tell me anything about the project that I would be working on but that it was important to the war effort and he was confident that I would find it interesting. I quickly accepted, with the request that I not be asked to build any more G-M circuits! I found out later that Wilma and Helen, who had married Glenn after my marriage to Wilma, had decided that I should join the Chicago group. Helen told her husband to hire Ghiorso and he took a chance and did so.

I was the twelfth member of the group and Glenn gave me a personal briefing when I arrived in Chicago a few months later in 1942. He told me in very broad terms about the atomic energy project, that their job was to determine the complete chemistry of an element that no one had yet seen. My job would be to take care of the instrumentation needs that his large group of chemists would use to do their job. He told me about the huge chain-reacting nuclear reactors that were contemplated to produce plutonium, the new element that he had discovered in Berkeley just two years before. And right next door to our laboratory near the University of Chicago, physicists were assembling the first man-made reactor in the world.

All of this information was overwhelming and it took a long time for me to grasp the enormity of the undertaking. It was very exciting to me and I, like everyone else, worked very hard. Six-day weeks were the norm and the necessary meetings that were held to keep the project on track were held at night so as not to disrupt the work. Glenn kept his promise — I did not have to build any new circuits; instead I had to maintain the ones that I had already built! This was in the days before transistors, when vacuum tubes had to be used for everything. The high mortality of the circuitry of the time was something that does not exist anymore.

From 1942 to 1946 I worked at the Metallurgical Laboratory, as it was known, learning the new arts of nuclear science. The work was fascinating and there was a lot to learn. The nuclear tools were rather primitive; hard careful work was necessary to gain the answers that we needed. For instance, to measure the energy of a gamma ray it was necessary to measure its rate of absorption in various materials by noting the decrease in the integral count as detected by a Geiger counter. Contrast this with the ease of making the same measurement in seconds with a modern germanium detector coupled to a multichannel differential pulse-height analyzer. It became particularly difficult when there was more than one component. It should be noted, however, that good work was possible even under these trying circumstances. For example, our discovery at Chicago of α -particle backscattering that I did under Burris Cunningham was deduced by careful integral counting of the activity emitted by uranium samples in a 2π α -counter. This effect was important, because it meant that the counting geometry for a weightless sample of plutonium in our 2π chambers was 52%, not 50% as had been assumed, and this affected the assays of how much plutonium was being used in a given experiment.

As time went on, it became clear that my chief value to the project would be not only to keep the equipment operating properly but also to assist in the development of new and improved methods of detecting nuclear radiations. I soon became the head of such a group and spent a lot of time doing experiments aimed at solving some of

the knotty problems concerning nuclear detection methods as they pertained to the research on the chemistry of plutonium. By 1944 Seaborg felt that he and his chemists had the chemistry of plutonium under control so that he could devote some time to looking for new elements beyond atomic number 94. He assigned me the task of developing a method for measuring α -particle energies with high efficiency and discrimination.

At that time the methods of making such measurements were very primitive and very inefficient. Library research soon showed us that no one had yet succeeded in that goal. We knew that one of the main problems which we would encounter was that, since it would be the target material in any nuclear bombardment and since the chemistry of the transplutonium elements was completely unknown, there would certainly be a large amount of ^{239}Pu in the final sample. I soon decided that one possibility that offered hope for discrimination from the plutonium α -activity was a range measurement, almost the equivalent of measuring energy. Since we were pretty confident that the range of the α -particles from element 95 or 96 would be longer than that of the plutonium alphas, the plutonium alphas would be absorbed before those from the new elements, leaving the new alphas by themselves without any background. In the past range measurements had been made at low geometry, thin absorbers being added incrementally to produce an integral range curve. I suggested that we might be able to make a good range measurement in the 2π ion chambers that we used for our regular assays of plutonium. The idea was to make the needed absorbers out of cleaved mica which would be very uniform in thickness and mount them on top of the sample to be analyzed. The end point of the absorption curve would be the range of the α -activity. Tests showed that the method was a satisfactory compromise for the work and we used it with notable success. The range method served its purpose for the discoveries of americium and curium, the first of the transplutonium elements to be discovered; a couple of years later the method was made obsolete by the invention of the gridded ion chamber by O.R. Frisch.

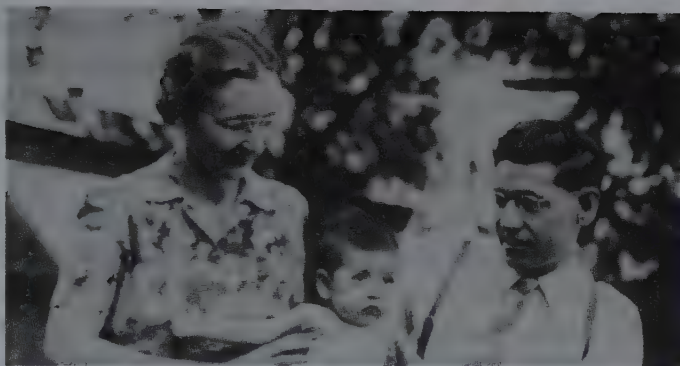


Fig. 2. My wife, Wilma, daughter Kristine, and myself, about 1945.

In 1946, with the War over, Seaborg decided to return to Berkeley to resume his job as a university professor and I was one of those invited to return with him. It was a difficult decision for me, as Wilma and I had become acclimated to the University of Chicago environment and set down roots. Our first child, Kristine, had been born there in 1944 (Fig. 2 is a photo of the three of us in 1945) and, believe it or not, we had come to even enjoy the City and its miserable weather. However, for me the career that I had embarked on as a scientist was too attractive for me to abandon and we, too, rode the train back to the West Coast to help set up a new laboratory.

Our son, William Belt Ghiorso, was born late in 1946. Thirty-two years later Bill was to become a member of the Laboratory, also, and join me in a notable experiment to produce element 110. My life changed in many other ways, too. Because of Wilma I became interested in classical music, art, and opera, and now, with more time available, we began to take advantage of the cultural vistas afforded by the Bay Region. In 1951 we bought our first modestly priced oil painting and that opened our eyes to the virtues of having original art as an important part of our surroundings, and about ten years later we started hanging some of our collection in my part of the Laboratory (Bldg. 71) to the delight of my colleagues. One of the great joys of the Berkeley region was its proximity to the high sierra

country and we soon began to spend our summer vacations camping in such places as Tuolumne Meadows in Yosemite National Park. In those early uncrowded days it was a wonderful place to relax and learn something about the wonders of the world around us and the important lessons of ecology.

As time went on and I gained more experience my role as an innovator became ever more important. The discovery of elements 95–101 was made possible by their chemical separation from the other elements utilizing Seaborg's actinide concept, but their actual detection was made possible by the development of new sensitive instrumentation and this is the area that I was to specialize in.

By 1950 another problem had arisen; we knew that at some point we would have to resort to particles heavier than helium to produce heavier elements and Seaborg suggested that it would be a good idea for me to explore whether the venerable 60-Inch Cyclotron could accelerate useful quantities of heavier ions. I did not know anything about the details of cyclotron operation, so I initiated a program of heavy-ion research by spending one day a week at that machine working with Bernard Rossi, who was in charge of the accelerator, to see if we could accelerate ions like ^{12}C . After a few months we were successful enough to be able to make several nuclear reactions in the transuranium region and show that a heavy-ion accelerator with more intensity and control of its energy was absolutely essential to our program if we were to go higher in atomic number.

Our success prompted Seaborg to request that a new accelerator be built at Berkeley for this research. Luis Alvarez, who had just invented the proton linear accelerator, suggested that Berkeley should build a linear accelerator for heavy ions and that it should make use of magnetic strong-focussing, a new principle that had just been invented at the Brookhaven National Laboratory. The AEC decided that two of them should be built, the other to be at Yale University. A study group was set up at Berkeley to design such a machine using scientists and engineers from both Yale and Berkeley.

The design effort went very well and in about a year the new linac was on the drafting boards and well into the initial phase of construction planning, etc., when an unforeseen stumbling block appeared. I attended the first Open House at the Laboratory, a joyous occasion as I remember, and everyone was there, including the Director, Ernest Orlando Lawrence. He took this occasion to ask me how the design of the new linear accelerator was going and said that he had been thinking about that machine and thought that it might be better if we built a cyclotron, instead! He suggested that a small group of interested people should get together the next day and discuss the idea. I was frightened by this proposal, because the design of our new machine was almost finished and the machine was ready to be built.

But this was E.O.L. himself, a very powerful individual and usually not thwarted, so the meeting was held. Ernest usually dominated technical meetings by the force of his personality and the fact of his many accomplishments, so it was not surprising that he would have an important influence in this battle of linac vs. cyclotron. I could see that the linac would go down to defeat unless it could be shown that it had some special advantage. It was a close call and the choice to a large extent depended on how one wanted to use the machine. Ernest was very enthusiastic about building a cyclotron, of course, and I could see that his point of view would carry the day unless I could point out something special about the linac. As the meeting went on I suddenly realized what that special advantage was.

At that time the problem of extracting the beam from a cyclotron had not been solved. The normal brute force method worked with very low extraction efficiency, a few percent usually, whereas in a linac it was obviously 100%. This argument was a powerful one and I kept playing this trump card whenever I felt that the cyclotron was winning out. The final outcome was that the linac design was allowed to go ahead. Twenty years later one could have made a better case for a cyclotron because of the development of the sector-focussed devices and ion source improvements, but at this time the

linac was certainly the best decision for Berkeley. If we had built a cyclotron, a chain reaction of later developments would have been precluded: the HILAC, the Omnitron, the SuperHILAC, the BevaLac, and RHIC at Brookhaven. Certainly, our heavy element program would have been delayed by several years. It is hard to know what the future would have been like; it certainly would have been different. After thwarting E.O.L. from making the HILAC into a cyclotron I thought that I would be *persona non grata* to him, but that was not the case at all. After the HILAC was built and operating he would often come around to find out how things were going, usually at night. He had the delightful habit of prowling the Hill at night occasionally to see who was working!

So construction proceeded and by 1957 the Berkeley machine (which I soon christened HILAC, for Heavy Ion Linear ACcelerator) was operating. The first experiment happened to occur on the same day that *Sputnik* was launched by the USSR. Thus was initiated at the same time the exploration of space by satellite and the exploration of the heavy element region by heavy-ion bombardment. Over the next ten years the HILAC allowed our team to produce for the first time elements 102–105. Element 106 would be made with the SuperHILAC.

While the HILAC design and construction was going on we exploited the 184-Inch Synchrocyclotron, using it to explore the region of elements between lead and uranium. The result was that a fascinating field was opened up, bringing to light a couple of dozen new nuclides as members of collateral α -decaying series. The information enabled us to study in great detail the systematics of α -radioactivity under the leadership of Prof. Isadore Perlman. In this work we took advantage of α -particle recoil to demonstrate the family relationships of the members of the series. Our research was then extended into the rare earths, where we found a whole new region of α -emitters.

In 1952 there occurred one of the most exciting incidents of my career, which culminated in the discovery of elements 99 and 100. The first hydrogen bomb test, conducted in the South Pacific by the

Los Alamos Scientific Laboratory, was analyzed by the Argonne National Laboratory and Los Alamos jointly and, initially, Berkeley did not even know about the operation. The amazing story of how we became involved is told in detail in Chapter 6 on einsteinium and fermium.

The bomb discovery of elements 99 and 100 was secret for a while and we began to worry that some other laboratory might find neutron-deficient isotopes of these elements by means of heavy-ion bombardment of uranium and, naturally, not knowing of our work, would want to name the new elements themselves. This would bring about a serious conflict, since we had already christened them einsteinium and fermium, so we decided to forestall this by finding these heavy-ion produced isotopes ourselves first. Since this work would not be classified, we would be able to publish it with a note that there was prior classified work on these elements which had priority of discovery. Our heavy-ion development at the 60-Inch Cyclotron paid off at this juncture by enabling us to find isotopes of 99 and 100 by ^{14}N and ^{16}O bombardment of ^{238}U . This work was published and it did accomplish its purpose of reserving the names einsteinium and fermium.

Within a couple of years we found that we could make the same heavy isotopes that had been produced in the Mike explosion by long and intense reactor neutron bombardments of ^{239}Pu . This material came from the so-called "napkin ring" bombardments in the MTR that had been initiated by Argonne and Berkeley around 1951. Micro amounts of einsteinium and fermium were now becoming available and we began to speculate as to how we could use this material to extend the periodic table.

For the elements beyond atomic number 100, however, conventional methods were not very efficient because of short half-lives and small amounts of activity. It became imperative that we develop new methods if we intended to climb up higher in atomic number. For the next element to be tackled, atomic number 101, I proposed that we bombard an unweighable target of ^{253}Es with a superintense beam of helium ions at the 60-Inch Cyclotron. Most important, I

suggested that we take advantage of the recoil produced by the transmutation process to separate the product atoms of element 101 from the target. It turned out to be very difficult to apply the principle because of the very low recoil energy but after we had overcome that problem by making very thin targets it was a very effective technique. This pioneering experiment of identifying a single atom at a time is described in Chapter 7.

To make the experiment possible Bernie Rossi, the Operations Chief, and I, working with the chief designer at the machine, "Chuck" Corum, had to make some major modifications to the cyclotron to obtain the necessary high beam density required for the experiment. Finally we were successful after many months of hard work and we were able to bombard our precious target of einsteinium with $10\ \mu\text{A}$ of He^{2+} ions in an area that was only a few square millimeters in size. I was so impressed by Corum that a few years later I hired Chuck to come and work with us at the brand-new HILAC. He was responsible for designing the complicated equipment that we needed for the heavy element experiments at that accelerator.

In October 1957, at Seaborg's insistence I attended the Materials Testing Reactor-ETR Users Meeting in Cincinnati, which would profoundly affect the future of the heavy element research effort. I had been asked to contribute a short paper on what the needs of our group might be in the future for larger amounts of transplutonium elements. Seaborg insisted that I go and "stake out our claim." Without thinking that it might ever happen, I gave a rather fanciful talk, extrapolating our needs for *multigram* amounts of the heavy curium isotopes and, as best I could, what might be accomplished with such generosity. My seat happened to be next to Dale Babcock of the Savannah River Project (SRP), where Du Pont had built and was running several huge nuclear reactors devoted to making *tons* of plutonium for the military program. After I gave my glowing pep talk in which I had plugged for a step-up in the production rate of the transplutonium isotopes by several orders of magnitude via either the ^{241}Am or the ^{239}Pu route, I sat down and Dale astonished me by saying that the Savannah River Project reactors could easily

produce the amount of material that I had been suggesting. All that would be required would be to have the MTR fuel assemblies refabricated to the one-inch diameter required by the those reactors and irradiate them further. I became quite excited and at the next break I sought out Bill Crane to tell him the news. Bill got his Ph.D. in Chemistry at Berkeley under Seaborg and had worked closely with us on a number of heavy element studies. After graduation he had taken a position at the Livermore Laboratory and now he was attending the meeting as their representative. He became enthusiastic also and the three of us joined forces in formulating a possible scenario and timetable for making macro quantities of americium and curium at the SRP. This would be used as target material for use in the projected High Flux Isotope Reactor (HFIR) at ORNL which, with a neutron flux of $\sim 5 \times 10^{15}$, would be able to make berkelium, californium, and einsteinium in large quantities.

The whole idea was quite stimulating to us. Here was the beginning of a grand new program that looked to be both scientifically and politically feasible. In fact, I was so taken by it that on the plane back home I wrote a memo to Glenn in which I espoused our proposed plan and delivered it to him the next day. The next step, Seaborg soon decided, was a letter to Lewis Strauss, the Chairman of the U.S. Atomic Energy Commission (AEC). In this October 1957 letter he recommended the need for a "very high flux reactor" (the HFIR which ORNL had proposed) and a twofold program to: (1) irradiate ^{239}Pu in a high flux production-type reactor (the SRR) to produce ^{244}Cm , and (2) irradiate curium in the "very high flux reactor" to produce berkelium, californium, and einsteinium in substantial quantities (milligrams!).

This was the beginning of a huge program that was to be so important to future developments in the heavy element field and it was going to take many years and cost many millions of dollars. Fortunately, Seaborg himself was to become the next AEC Chairman, so he was in a perfect position to oversee it and make sure that the program stayed on course. And that it did. The HFIR was built and operating by 1965. In addition, the Transuranium Processing Plant

(TRU), needed to process the transplutonium products from HFIR, was built and operating by 1966.

The National Transplutonium Program was a cooperative one, aimed at benefiting all the laboratories in the US, so in 1964 the Transplutonium Program Committee was set up with members from the principal laboratories involved in heavy element research — Argonne, Berkeley, Livermore, Los Alamos, and Oak Ridge — to advise the director of AEC's Division of Research on how the actinide products should be allocated. There was usually an equal division of the production to the five principal members: but that was often modified when a particular laboratory had a special experiment which needed all of the current production. I served on the Committee continuously until it was disbanded. I recall asking the Committee in 1967 for the allocation of *all* of the first batch of pure ^{249}Cf (amounting to several hundred micrograms) that would be milked from the ^{249}Bk "cow" which had been purified and set aside for the purpose of growing ^{249}Cf . I told the Committee that it was to be used in our initial attempt to make element 104. The Committee, under the capable leadership of first Alexander Van Dyken and then John Burnett, was very proud when I was able to report back to them a year later that we had succeeded. Van and John were very popular with the members for the wise roles that they played in satisfying the varied, sometimes conflicting, interests of the members during the many years that the Committee existed. Often, in the early days of the Program when the actinide element shares were meager and the competition between the laboratories was keen, there were lively debates over where the "goodies" should be shipped first. These disputes were always settled amicably by cooperative agreements and a trading of priorities, the guiding rule being that the materials should be used to produce the best science.

The equipment needed for us to discover elements 104, 105, and 106 became more and more complicated, of necessity, because the bombardment yields decreased steadily as the atomic number increased. Fortunately, the development of solid-state detectors made it possible to design experiments which were marvels of sensitivity.

The last of these, for element 106, was even able to demonstrate the presence of the great granddaughter of $^{263}\text{106}$. The apparatus had the designation "VW." This stood for "Vertical Wheel," a descriptive term for an apparatus with unparalleled sensitivity at the time. It was the culmination of a line of instruments that identified α -particle activities by their α -energies and by the genetic relationships that they had to other α -activities. With this instrument we were able to refine our research and characterize isotopes with great accuracy, even when there were interfering activities present.

Though we specialized in α -emitters we did not neglect those nuclei that decayed by spontaneous fission. G.N. Flerov and his group emphasized SF emitters from the beginning of their foray into the heavy element field and continued to make claims that had to be contended with, starting with element 104. It was necessary for us to find out whether their claims were correct, so we gradually devoted a fair amount of time to checking them. We soon found out that it was not possible for anyone to be absolutely certain of the isotopic or element assignment of an activity of this sort with the exception of the few times when it was possible to carry out well-established chemical procedures. We soon let it be known that we considered that one α -particle was worth at least ten thousand fissions! However, we did much research in this area under the tireless leadership of Matti Nurmi and were able to find several important isotopes that decayed principally by spontaneous fission.

Two very important events happened during this period. Iz Perlman had asked me if I would watch over the HILAC and I did. Soon, however, I found that the actual technical job took too much time away from my research, so I hired a young man, Bob Main, away from TracerLab to take care of these day-to-day duties. That was an excellent move, because Bob (see Fig. 3) was not only an excellent physicist and engineer, he was also a businessman and that skill soon became very important as we got involved in the design of new machines. Though I no longer had to watch over the detailed operation of the HILAC, I remained interested in accelerators in general and I would often discuss fine points with Bob. Thus,

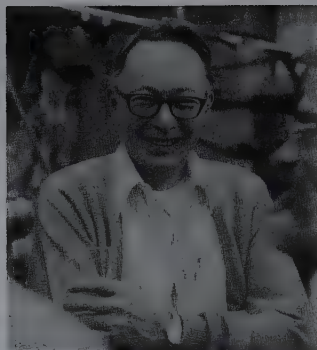


Fig. 3. Robert Main, in charge of the operation and development of the HILAC and the SuperHILAC. Coinventor of the Omnitron.

when Glenn, who had become the Chairman of the Atomic Energy Commission in 1961, informed me in a chatty letter from Washington that the Argonne Laboratory was thinking of building a large cyclotron for heavy ion acceleration, I took it as a challenge and began to ponder what we might do for Berkeley's future. I posed the problem to Bob and found that it was a very expensive proposition to make a magnet with a pole diameter of the order of 200 inches! We were thinking of a magnet that large so that we could take ions out of the HILAC up to higher energies principally for the use of the biomedical community; and with such a huge magnet we knew that we would be able also to accelerate the abundant low charge state ions to get large beam currents of ions with energies suitable for making nuclear reactions. Bob had invented a new method of winding magnet coils with heavy copper tape which promised great savings, but a careful study quickly uncovered severe problems and we abandoned that approach. Once we had got our feet wet in the field of heavy ion accelerators there was no turning back and we returned in our thinking to the cyclic machines, still guided by the desire to accelerate high energy heavy ions for biomedical usage.

There were a few more iterations in our thinking until one memorable hour in 1964 when Bob Main, Bob Smith, and I invented a new

type of accelerator which we called the Omnitron, one which could accelerate all of the elements to either low or high energies. It was a real breakthrough, far ahead of its time, and was one of the world's first complicated accelerator concepts. This machine would have accomplished its purpose by the use of two large synchrotron rings of magnets in which the particles were accelerated and/or stored. The particles could be passed easily from one ring to the other so that a cyclic regime could be set up in which particles could be accelerated first in a low and then in a high charge state. We even pointed out that radioactive ions made by fragmentation could be accelerated efficiently — a modern concept. If we had built the Omnitron there is no question but that the history of the Lab would have been changed drastically, because many powerful tools would have become available two or three decades before their time.

The other event of great importance was the publication in the same year of the seminal work of Bill Myers and Wladek Swiatecki which suggested that a possible closed neutron shell at 184 neutrons and one at 126 protons could lead to a region of very stable super-heavy elements (SHE). Some of the best combinations of elements needed to make the SHE were of high atomic number, so it became immediately clear that our proposed machine, the Omnitron, would be the ideal accelerator to implement the fusing of the necessary atoms to reach this Magic Island. However, our original reason for inventing the Omnitron was to accelerate heavy ions such as neon to energies high enough to penetrate the human body for the treatment of cancer.

Fortunately, Seaborg was the Chairman of the AEC and he was definitely interested in our pursuing the search for SHE, so it was not difficult to get him interested in the capabilities of our novel machine. Funds were soon made available for us to do a design study and this led to a full-fledged proposal to construct the Omnitron. Figure 4 shows an architect's model of the Omnitron superimposed on the hill area near the 184-Inch Cyclotron. The study, very carefully done by a group of the best accelerator people in the US, concluded that the Omnitron would have a construction

cost of \$28 million but operating costs that were quite low, only about \$3 million/year; it was deemed to be a highly worthwhile project. Twenty-five years later the design was found to be still excellent and not at all outdated. Figure 5 is a photograph of the author at about this time.

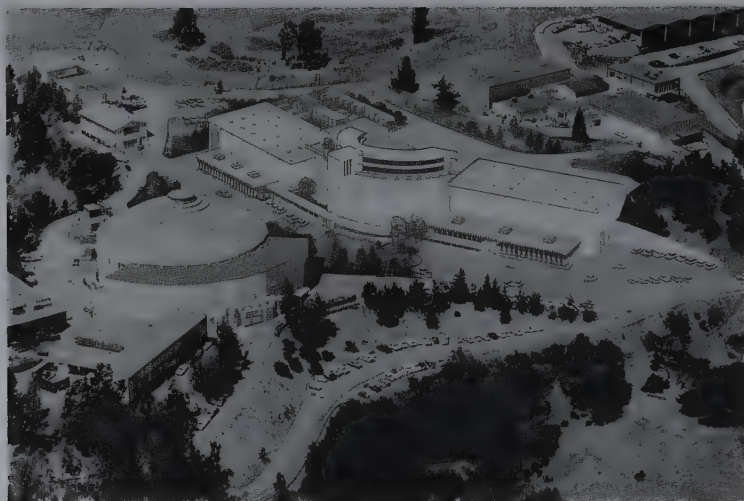


Fig. 4. Architect's drawing of the Omnitron model superimposed on the hill area near the 184-Inch Cyclotron.

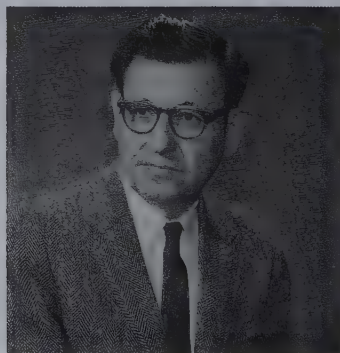


Fig. 5. Ghiorso, approximately 1969.

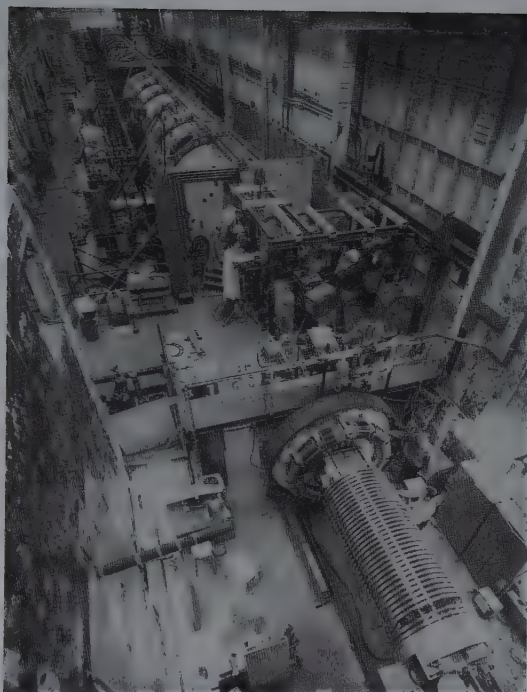


Fig. 6. The SuperHILAC in the final stages of construction, 1971. Some of the iron shielding is not yet in place. The ADAM injector is at the lower right.

Unfortunately, the accelerator was never built because it came into competition with the terrible holocaust war in Vietnam, where the US was spending the equivalent of three Omnitrons per day to devastate the country! We had prepared a “fallback” position in case we failed to get approval, in the form of important improvements to the HILAC that would cost only \$3 million. We were able to get these funds and the SuperHILAC came into being in 1971. Figure 6 is a photograph of that machine in the final stages of construction.

In retrospect, I think that we pursued the wrong strategy. I now believe that we should have continued to press for the Omnitron. There is no question that we would have failed to get approval for

the project in the following year or two, but it was clearly a superior accelerator with no competition in its field and I firmly believe that eventually it would have won out. It was 25 years ahead of its time and would certainly have changed the course of nuclear physics at LBL.

Though not as versatile as the Omnitron, the SuperHILAC was a great improvement over the existing HILAC and with it we were able to pursue our heavy element research. It was with this machine that we were able to discover element 106 in the form of $^{263}106$ in 1974.

In 1971 the BevaLAC was conceived. At the time this was a startling concept, and it came about in this way. I had felt guilty that we had not been able to build the Omnitron for the biomedical community and kept promising Cornelius Tobias and John Lawrence that I would find some way to increase the energy of the heavy ions that would come from the SuperHILAC. At first, we thought in terms of just adding more linacs, but it became clear that this would be impossibly expensive. However, one day when I was pondering a layout of the Lawrence Berkeley Laboratory that showed all of the accelerators on the Hill, I noticed that the Bevatron in plan view seemed very near to the SuperHILAC. Somewhat flippantly, I made what seemed like a good joke by saying that we ought to inject our SuperHILAC beams into the Bevatron! It seemed like such a novel idea that I thought we had better calculate why this was not feasible, but a few minutes of calculation by Frank Selph showed that it could be done. Figure 7 is a schematic diagram of the BevaLAC arrangement. It was thus that the BevaLAC was born and on the spot I coined that acronym. With Seaborg's help in securing funds from the AEC (he had just completed his ten-year stint as Chairman in Washington and had returned to Berkeley) the necessary transfer line was constructed to connect the two accelerators, the BevaLAC came into being and my promise to John Lawrence was fulfilled. It was very successful and after a learning period of several years it was shown to be an important biomedical tool. In addition, interest was aroused in using it as a probe to form ultradense matter, the

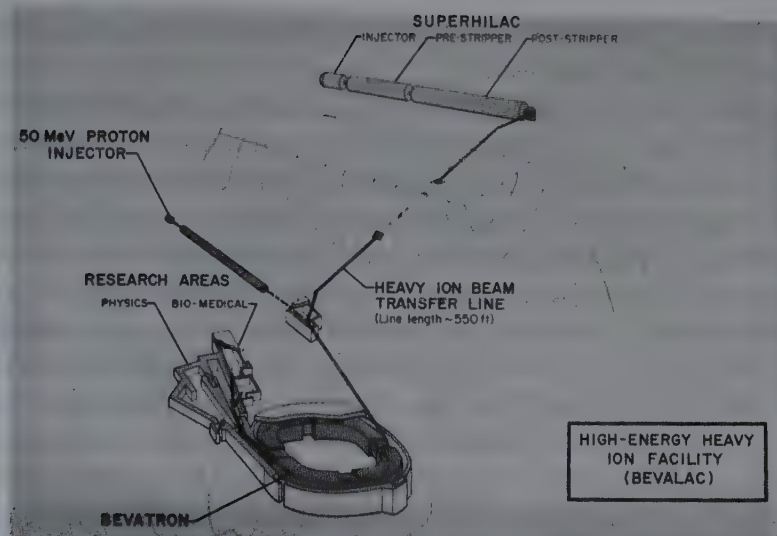


Fig. 7. Schematic diagram of the BevaLAC arrangement.

famous *quark-gluon* plasma which must have existed for a brief instant at the time of the big bang.

This early work with the BevaLAC aroused a lot of interest among the high energy nuclear physicists and after a few years prompted the development of a heavy ion capability at the existing large hadron accelerators to achieve much higher energies. No quark-gluon plasma evidence was seen and it was realized that even higher energies would be needed. This insight led to the construction of the heavy ion collider, Relativistic Heavy Ion Collider (RHIC), at Brookhaven to go in the existing tunnel that had been constructed there for Isabella, a high energy proton accelerator that was never built. Thus, it is evident that the BevaLAC not only kept the Bevatron going for another 20 years, it also opened up a completely new field of physics and thus had a very great impact on worldwide research. The irony of it is that when the Bevatron was shut off in 1993 the SuperHILAC was also shut down as its injector, even though there were many important experiments that could have been done at the

SuperHILAC that could not be performed by any other accelerator in the world at that time.

My promise to the biomedical community was fulfilled, but what about the low energy uses that the Omnitron was designed to handle? The SuperHILAC was meant to accelerate copious beams of heavy ions with atomic number up to 36, i.e., krypton. This usage of the machine had many growing pains and though it was designed to work in a time-sharing mode with three different injectors so that one beam could be injected into the Bevatron while a different beam could be used for low energy experiments at the SuperHILAC, it was very difficult to accomplish this all of the time. It seemed that the highest priority usually went to the Bevatron, since it was the most expensive part of the complex to operate.

However, some of the time it worked very well for us. We were able to mount a very excellent experiment in 1974 in which we discovered element 106 by bombarding ^{249}Cf with ^{18}O ions (see Chapter 10). This was done following many different types of experiments aimed at making the longed-for superheavy elements. None of them worked and in retrospect it was mostly time wasted that could have been devoted to more productive research. But there was no way that we could foresee that this would be the case! When we finally tried the element 106 experiment, we were on more familiar ground which was an extension of what we had been doing for years.

During the period that the Omnitron was under development by a corps of some of the best accelerator physicists in the world who had been recruited by Bob Main, I followed the work with great interest and as a result came up with an excellent idea for a new kind of separator. I was struck by the fact that a fast-moving ion of a heavy element that emerged from a foil into vacuum had a higher mean charge state than when it emerged into a region of low gas pressure, and furthermore, the charge state attained depended on the atomic number of the ion. Might this be a way of separating a heavy ion recoil from the beam that produced it?

I was struck by the simplicity of the idea, so I did a simple experiment at the exit of the HILAC in April 1967, before it was

converted into the SuperHILAC. I mounted a thin target of ^{165}Ho at the entrance to the steering magnet at the end of the accelerator and bombarded it with a beam of ^{40}Ar ions. An aluminum catcher foil was mounted downstream at the end of the magnet so that an α -radioautograph could monitor how much the 40 MeV α -emitting astatine recoil products of the bombardment were bent relative to the argon beam by a given magnetic field when the pressure was about 1 torr of helium. An excellent separation was obtained. A later experiment using 67 MeV ^{12}C ions to bombard a gold target also worked extremely well, with the 4 MeV astatine recoils also being completely separated from the beam. In addition, it was noted that a sharp image of a collimator was obtained and this indicated that charge-exchange oscillations occurred very frequently along the recoil trajectories so that the magnetic rigidity was constant. I learned somewhat later that my work had been preceded by Fulmer and Cohen at the Oak Ridge National Laboratory when they applied the principle to the separation of fission products recoiling from a fission source.

Unfortunately, this promising technique was not adequately followed up until much later, because the successful experiments on elements 104, 105, and 106 took precedence and they used our more conventional techniques. It was not until 1972 that our first steps were taken to make a gas-filled recoil separator that would be suitable for work with heavy ions. A device called SASSY (Small Angle Separator System) was constructed using available magnets from the Bevatron. It had some good ideas, such as the measurement of the time of flight (TOF) of the recoils, but suffered from the fact that the discrimination at the focal plane from beam particles was insufficient at high beam levels. In addition, the silicon detector array was not large enough to catch all of the fusion recoils.

Some years later another version of the idea was constructed, called SASSY2, which remedied these faults. It had a larger acceptance for the recoils and these were bent through a much larger angle. By this time I had "retired" and so had no access to funds which would have allowed the instrument to be constructed in the

normal way. Faced with this situation, I had no choice; if I wanted to pursue the heavy element research in this way, I would have to build it myself. I had often built parts of the instruments that I used but now I would have to build the major parts of SASSY2. For instance, the pole pieces would have to be milled in a complicated way to provide the necessary double-focussing. This was a daunting task but my son, Bill, persuaded me that it could be done and offered to help me with both advice and labor. He had joined the Lab about ten years before and had become an acknowledged expert in mechanical, electrical, and computer technology. His help and enthusiasm were prime reasons for the successful operation of SASSY2 and we did succeed in making a viable instrument that was used for one important last experiment. This was a search for element 110 with mass 267 produced by bombardment of ^{209}Bi with ^{59}Co ions. In a very difficult 40-day period we did find one event which we attributed to this atom and that experiment is described in Chapter 12.

Subsequently, the SuperHILAC was shut down, prematurely ending our encouraging experiments there. However, SASSY2 was especially important in that it led directly to the design and construction at the 88-Inch Cyclotron of the Berkeley Gas-filled Separator (BGS), the next generation of gas-filled separators that came on line early in 1999. (See Chapter 14.)

With this device we were able to mount an experiment to look for the isotope, $^{267}110$. In a very difficult 40-day period we did find one event which we have attributed to this atom. The experiment is described in Chapter 12.

P.3. Glenn T. Seaborg

I had almost no exposure to science in my early years. I was born of Swedish ancestry in Ishpeming, Michigan, a small iron-mining town on the Upper Peninsula. My father, H(erman) Theodore Seaborg, was born in 1880 in Ishpeming. His parents came from Sweden to Ishpeming in their youth and met and married there in 1872. His

mother, Charlotta Wilhelmina Johnson (whose family name was changed to Farrell), came to Ishpeming in 1869 at the age of 19 from Örebro with her parents and brothers and sisters. His father, Johan Erik Sjöberg (whose name was anglicized to Seaborg), came to Ishpeming in 1867 from Hällefors at the age of 23. Johan's father, Erik Sjöberg, changed his name from Olsson in 1835. As I recall my father telling me, Johan crossed the Atlantic Ocean as a steerage passenger in a cargo ship. Johan had a friend at the Hällefors Iron Workers, the grandfather of the Swedish Nobel Prize winner, The Swedberg, so I suspect that the name The and my middle name Theodore have a common origin.

The Seaborg home in Ishpeming at 639 East Division Street was occupied by members of the Seaborg family until 1914, then by members of the Kurin family until 1980, when I acquired it, and I still retain it for sentimental reasons.

My mother, Selma Olivia Eriksson (changed to Erickson), was born in Grängesberg in the southern Dalarna region of Sweden, and came to the United States (Ishpeming) in 1904, when she was 17 years old. She and my father met at a picnic on Swedish Midsummer's Day, June 24, 1908, and were married three years later on Swedish Midsummer's Day, June 24, 1911. I was born in Ishpeming on April 19, 1912 (during the presidency of William Howard Taft).

My mother's ancestors had lived in the southern Dalarna–northeastern Västmanland region of Sweden for many generations. A home in which her ancestors, Michael Hindersson and his wife, whose maiden name was Maria van Gent, lived in 1673 (as shown by an inscription on the living room wall) was moved in 1895 from Kopparberg to Skansen, where it stands with the name "Laxbrostugan" as part of the representative houses from "Bergslagen." This house has served as the rallying point for hundreds of my relatives, and has given me contact points to enable me to trace my ancestry back to the 14th century in Holland (Maria van Gent's birthplace). The Hindersson's daughter Britta married Nikolas Pemer, whose family came from Augsburg, Germany. The Pemer clan has formed a "Pemer Society" in Sweden in which Helen and I and our children are among

the hundreds of members. Here my ancestry has been traced back to the 15th century.

Ishpeming had typical sections that were nearly all Swedish and it was in one of these that we lived. Since my father was fluent in Swedish and this was my mother's native tongue, the Swedish language was spoken in our home as it was throughout the community. I learned to speak and understand Swedish before I did English, but I am afraid that in the intervening years my facility with the language has declined. My younger sister Jeanette also spoke Swedish as her first language.

I was born in the second story rented home of my parents at 231 New York Street, in a section of town called the "Old Location," named after a nearby abandoned iron mine. (This old house is still there in 1998.) The congested "Tangletown" neighborhood, so called because of the mixed-up labyrinthine nature of the streets, contained the homes of our Swedish relatives and friends — the Swansons, Swanbergs, Hedstroms, Bjorks, Petersons, Greens, Quaals, Samuelsons, Olsons, and Dahls. When I was three and sister Jeanette one year old (Fig. 1), my parents purchased and moved into a two-



Fig. 1. Childhood picture of Glenn T. Seaborg with his sister Jeanette, ages four and two, Fall 1916.

story house about a quarter-mile north of the Old Location at 802 East Wabash Street, at the corner of 7th Street. (This old house is also still standing in 1998.) Ishpeming had severe winters with high levels of snow. I recall climbing out of my second-story bedroom window with my skis strapped on for an adventurous cruise on the top of the snow pack at near the house top level.

Here my main playmate was Clarence Larson, a classmate who lived diagonally across the street. Clarence had a younger brother Raymond who was about Jeanette's age and her (and my) close friend. My other playmates included Laurel "Dirt" Williams, Clarence "Cuckoo" Vinge, Ralph Haugland and his younger brothers "Winky" and "Coonjigger," Carl "Issy" Carlson and his sisters Anna, Esther, and Margaret (about my age and a classmate), Toive Dahl, and Eric Dahl. Most of us had nicknames. My nickname was "Lanky," an obvious appellation (a third-grade picture shows that the tallest of my classmates only came to my shoulders) (Fig. 2). On a visit to Ishpeming in 1994, I called on Dirt Williams and found him living in the same house that his family occupied when I lived there more than 70 years ago. I have kept in touch over the years with Clarence "Gom" Larson, who has lived in the neighborhood of Ishpeming throughout his life.



Fig. 2. Third Grade Class, High Street School, Ishpeming, Michigan, June 13, 1921, showing Glenn T. Seaborg as the tallest student in the back row.

Swedish customs of all kinds prevailed in our home. I remember particularly well the Swedish food that we enjoyed at our dinner on *Julaften*, or Christmas Eve. The fare usually included *smorgasbord*, which featured *sil*, or pickled herring. One of the mainstays was *lutfisk*, which was always served with boiled potatoes and a white sauce. Another feature was saffron buns and bread, usually served hot and made with glaceed fruits. This was part of a large spread of buns and cakes, including *gingersnaps* made in the form of goblins, piglets, stars, and other patterns. Another component which was almost always present was the Swedish *lingonberries*, which I still like so much. The meal was usually rounded off with *risgryn*, or rice pudding, which was topped with cinnamon with cream and sugar. Even in later years my mother carried on these traditions and my wife, Helen, has done her best to continue such activities for the benefit of our children.

Ishpeming during my first ten years was an isolated world of its own. I never saw, or even heard, the word "radio." I don't recall speaking into a telephone. We were served by a weekly newspaper, *The Ishpeming Iron Ore*. The dirt streets had a fringe of red color due to the iron ore "*hematite*," which topped underground deposits laced with miles of tunnels for the mining operations. We did have some access to nearby Marquette's daily *Mining Journal* and to the Sunday edition of the *Chicago Tribune*. I recall reading the Sunday comics featuring classics such as "The Katzenjammer Kids," "Slim Jim," and "Bringing up Father" (featuring the irrepressible Jiggs).

I started kindergarten in the High Street School in September 1917 and continued there through the first three grades. For the fourth grade I moved in 1921 to the Old Grammar School a couple of blocks west (corner of First Street and North Street). In the third, fourth, and fifth grades I was an ardent admirer of a girl named Dorice Gray. During the summers of 1920, 1921, and 1922, when I was eight, nine, and ten years old, I worked as a caddie at the Ishpeming-Negaunee nine-hole golf course (the Wawonowin Golf Club), where caddie rates were 20¢ for nine holes, paid by one category of player, and a 25¢ charge for the more affluent players. We sometimes

maintained the Ishpeming or Negaunee gate (opening the gate for an incoming automobile). A small coin (a penny, nickel, or sometimes a dime) was thrown on the ground for us. Dorice's father was the manager of the golf club, and thus I was able to admire her from a distance during my summer visits to the Club House area.

Although I had become sufficiently fluent in English to cope by the time I started kindergarten in the fall of 1917, I was so shy as to cause problems. My mother had to negotiate a special arrangement with the teacher, Mary Earle, to allow me to go directly to the restroom during class time, without having to raise my hand and speak up to ask permission (an act that was beyond my capabilities).

The flu epidemic struck Ishpeming, as it did throughout the country, in the fall of 1918. My parents and many of my neighbors were afflicted and I recall that a number of our friends succumbed. I also remember vividly being surprised that I was so sick when my turn came. My sister Jeanette's flu turned into pneumonia, which gave us great cause for concern until her recovery.

In 1922, when I was ten years old, my family, which included my younger sister Jeanette, moved to Home Gardens, now a part of South Gate, California (near Los Angeles). At this time I changed the spelling of my name from "Glen" to "Glenn." This move was made largely at the urging of my mother, who wanted to extend the horizon for her children beyond the limited opportunities available in Ishpeming. However, unlike in Ishpeming, where he would have had guaranteed employment for life, my father never found permanent employment at his trade in California, and our family found itself in continuing poor circumstances. Since the new subdivision of Home Gardens had no schools, my sister and I during the first year traveled by bus to attend the Wilmington Avenue Grammar School in the Watts district of Los Angeles. I completed my grammar school education through the eighth grade in the newly constructed Victoria Avenue Grammar School in Home Gardens, skipping a couple of semesters on the way to my eighth-grade diploma.

When I entered David Starr Jordan High School in the Watts district of Los Angeles, again traveling to school by bus, I had to

choose between a commercial and a college preparatory curriculum. My mother pressed for the commercial course; to her this was the road to a respectable white-collar job. But I started down a different road and chose the college preparatory program, with literature as my major subject. During my freshman and sophomore years, I studied the usual college preparatory subjects, such as English Literature, Oral English, and World History, as well as Algebra, Geometry, and a foreign language.

In my junior year I was required to take a laboratory science in order to be eligible for admission to the "tuition-free" University of California at Los Angeles (UCLA). Because my high school was small, Chemistry and Physics were offered in alternate years, and Chemistry was the offering in my junior year. It was fortunate for me that my first science course was taught by Dwight Logan Reid, an outstanding teacher who exerted a strong formative influence on me. Mr. Reid not only taught Chemistry, he preached it. He related some fascinating experiences he had had as a Chemistry student in college, and, when he lectured, his eyes would light up. His irrepressible enthusiasm, obvious love for the subject, and ability to inspire interest captured my imagination almost immediately. Early in his course I decided I wanted to become a scientist. As a senior I took Physics, also from Mr. Reid, and since then my interests in Physics and Chemistry have been inseparable.

Immediately after starting at Jordan High School, at the age of 13, I fell madly in love with Vivian Dawson, a slim brunette of greater than average height with flashing eyes that radiated intelligence, a fellow freshman, and a resident of Watts. We had adjoining seats in the Oral English class, which gave us a chance to communicate and get well acquainted. I believe she was also attracted to me. She shared her chocolate bars with me and was very friendly. I was too shy and inexperienced to take advantage of the opportunities to walk her home after school and after football games — stupidities that I have often, in retrospect, regretted. She left Jordan High School in the middle of the semester, when her family moved from Watts. I don't know where she went and I never saw her again. I have

always hoped that we would meet again to bring each other up to date on our subsequent activities.

Early in 1927 I saw at a basketball game Bonita Edwards (an eighth-grader) and was struck by her vivacity and brunette beauty, and her resemblance to Vivian Dawson. Although shorter than Vivian, she also had sparkling eyes that advertised intelligence. (She graduated from high school at the age of 16.) She lived in Watts, across the street from Jordan High School. To my delight in the fall of 1927 as a freshman, she was a member of Charles Hicks' Latin I class, in which I was also enrolled as a junior. (Her attractive sister, Claire, a sophomore, was also a member of this class.) During the following two years of Latin class, I had an opportunity to get well acquainted with Bonnie and Claire. I became enamored with Bonnie but was too shy to advance my cause. I was ecstatic when, in the spring of 1929, she asked permission to wear my senior class ring. I was devastated when she returned to me my ring at the end of the semester, as she had, unfortunately, promised to do. Again, in retrospect, I have often ruminated on how clumsily I handled this situation. Bonnie married a football-playing friend of mine, Bud Coffin, and we have been lifelong friends.

After I finished high school in June 1929, I was very fortunate to find employment during the summer as a laboratory assistant, working as the lone control chemist on the graveyard shift (11 p.m. to 8 a.m.) at the Firestone Tire and Rubber Company in their South Gate plant. This provided the money that made it just possible for me to enroll at the University of California at Los Angeles (UCLA) in the fall. I knew I wanted to major in either Physics or Chemistry. I believed that a physicist could make a living only by teaching in a university, and at that time university faculties had few openings. On the other hand, a chemist unable to find a university teaching position could go into industry. So I chose Chemistry, hoping to become a university teacher, but, knowing that if I did not, other career opportunities would be available.

I lived at home and commuted by car with Jordan High School friends a distance of some 20 miles to UCLA. I have continued

lifelong friendships with many of my classmates at Jordan High School in Watts. Stanley G. Thompson, who lived in Watts with his grandmother, was a sort of boisterous roughneck when I first encountered him in the ninth grade at the age of 13. He became a serious student when he reached the Chemistry class in his junior year and received the top grade in the second semester. We attended UCLA as Chemistry majors, traveling on occasion in his new Ford sedan, purchased for him by his "aunt" Bessie Brigance (who, I learned later, was actually Stanley's mother). We retained a close relationship before and during our wartime service on the Plutonium Project at the Metallurgical Laboratory at the University of Chicago, and then at the Lawrence Berkeley Laboratory (LBL) at the University of California (UC), until his death in 1976. (I had the pleasure of serving as best man at his marriage to the delightful Alice Smith, a San Diego girl, on Sunday, November 27, 1938, at Northbrae Church in Berkeley.) Stan was an extraordinarily able chemist with the best intuitive sense, *Chemisches gefühl*, for solving chemical problems of anyone I have ever known. As I shall recount later, he solved the problem of chemical separation of plutonium on the Plutonium Project. We were collaborators on the synthesis and identification (i.e., the discovery) of a number of transuranium elements.

One sunny morning in September 1929, I walked across the ravine on the bridge which served as the entrance to UCLA from the Hilgard Avenue (east) side in Westwood. This was the opening year on this site of this young campus, only ten years old. Los Angeles State Normal School, on North Vermont Avenue in Los Angeles, became the Southern Branch of the University of California in 1919 and then the University of California at Los Angeles in 1927. There were some 5000 students and the total facilities at the opening of the Westwood campus in 1929 consisted of four buildings and a couple of temporary structures, including a student bookstore and gymnasium facilities. The four buildings, situated in a quadrangle, were the Chemistry Building (now Haines Hall, housing Geology), the Physics-Biology Building (now Kinsey Hall for Physics), the

Library Building (now Powell Library), and Josiah Royce Hall (named for the famous American philosopher and accommodating the other departments, including Mathematics) (Fig. 3). We found these not-quite-completed buildings rising starkly from the bare earth. Raw lumber and sacks of cement lay stacked for use. Lawns and shrubbery were nonexistent and the dusty walks turned to lanes of gooey mud when it rained.

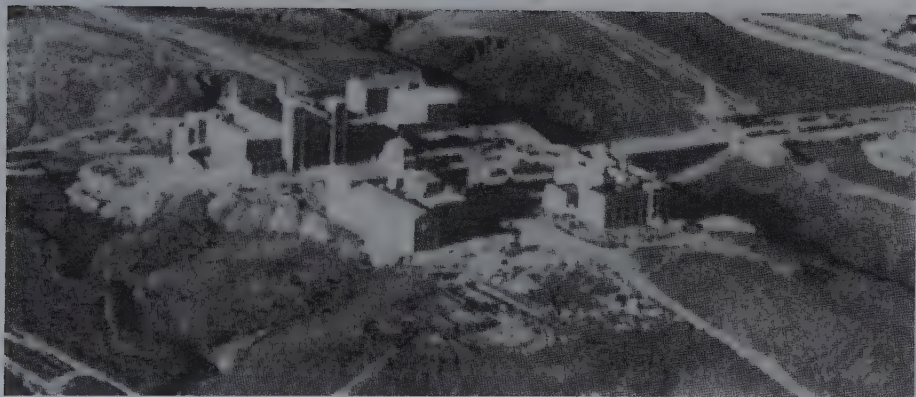


Fig. 3. UCLA campus and environs showing Library, Royce Hall, Chemistry Building, and Physics Building, 1929.

I recall vividly the first meeting of my freshman chemistry class in the fall of 1929 with Prof. William Conger Morgan, a formidable man, some six-and-a-half feet tall. When Prof. Morgan strode into the auditorium of the Chemistry Building he glowered at the 300 students filling the room. He finally broke his silence to announce in a stentorian voice, "Look at the student on your right." After we had all done this, he commanded, "Look at the student on your left." After we had all done this, he bellowed, "One of you three will not be here at Thanksgiving time." I resolved to survive past his deadline.

In 1929, UCLA offered unique opportunities for undergraduates in Chemistry. In addition to Prof. William Conger Morgan, chairman of the Chemistry Department, Profs. William R. Crowell, G. Ross

Robertson, J. Blaine Ramsey, Hosmer W. Stone, Max S. Dunn, and a year or two later, William G. Young and Francis E. Blacet offered an extraordinary curriculum. The absence of graduate work in those years was probably the reason our able professors gave us a taste of graduate-type research by the time we had reached our sophomore or junior years.

At the end of the fall semester, in January 1931, I received a grade of 99% in the final examination for Prof. William R. Crowell's class in Quantitative Analysis (Chemistry 6A). This so impressed Prof. Crowell that he gave me the job to serve one afternoon a week as laboratory assistant in his Quantitative Analysis course for premed students (Chemistry 5). He also set me up in a job one afternoon a week in the stockroom for the freshman chemistry laboratory (Chemistry 2A and 2B for nonmajors), checking out to students chemicals, equipment, etc. These jobs, a total of six hours a week at the magnificent pay of 50 cents per hour, restored me to financial solvency. To add to my security, I received in May a letter from UCLA Recorder H.M. Showman, advising me that the UCLA Committee on Scholarships had granted me a \$150 scholarship for the academic year 1931–1932.

One day in the summer of 1932, Stan Thompson and I paid a nostalgic visit to the Long Beach home of our high school chemistry teacher, Dwight Logan Reid. Here I met his attractive daughter Beth, about a year younger than I, with whom I was very much impressed. She had attended two years of junior college in Long Beach. I was delighted when I met her about a month later, when she came to register at UCLA as a junior with a major in Physical Education. Although much interested, I didn't have my first date with Beth until the following spring, when I escorted her to a party of my professional chemistry fraternity, Kappa Gamma Epsilon, which I was serving as president (and which I later led into membership as the Beta Gamma chapter of the national professional chemistry fraternity, Alpha Chi Sigma). Over the next year or so I had dozens of dates with her — escorting her to parties, playing tennis, accompanying me to football games at the Los Angeles

Coliseum, attending noontime assemblies in the auditorium of Royce Hall, etc. We attended a "pajamarino" celebration at a bonfire on the lower UCLA campus the night before the first-ever football game between UCLA and Cal-Berkeley. Beth and I were in the Los Angeles Coliseum the following afternoon to witness the historic 0-0 tie and I witnessed the ceremony just before the kickoff when UC President Robert Gordon Sproul, student body presidents Wakefield Taylor (Berkeley) and Porter Hendricks (UCLA), and graduate managers Bill Monahan (Berkeley) and Bill Ackerman (UCLA) met on the Coliseum turf. My dating of Beth ended when I left for Berkeley in the fall of 1934, but we have remained friends and seen each other on many occasions in the intervening years.

I have an especially vivid memory of the Friday, March 10, 1933, widespread earthquake centered in the Long Beach area. Stan Thompson and I were driving home from UCLA in his Ford along Slauson Avenue when the earthquake struck at 5:55 p.m. The sensation was so severe we had to stop driving. The Huntington Park High School was on fire when we drove past and we saw many demolished buildings as we approached home. When we arrived home in South Gate my mother and father and sister Jeanette were out in the front yard, having evacuated the house. I went in our house, found our dinner (pork chops) all over the floor in the kitchen, and furniture throughout the house overturned (i.e., the radio in the living room; the typewriter which normally sat on a table in my front bedroom had been thrown over onto the bed). I slept in my Chevrolet coupe that night, and my parents and sister slept in the family Star car. The next day I drove around to view the earthquake ruins. Compton and Long Beach were hit worst, with Watts, Huntington Park, Huntington Beach, and Santa Ana also hit hard. The morning newspaper estimated the death toll at 127, with 3000 injured and \$30 million worth of property damage. Tremors continued throughout the day and the next day. However, when I returned to UCLA on Monday I found very little earthquake damage there.

While majoring in Chemistry, I took the maximum number of courses in Physics. In my senior year at UCLA I had a course in Modern Physics given by Prof. John Mead Adams (a lineal descendant of the second president of the United States), who talked to us of the exciting discoveries in nuclear science. And these lectures fixed my sights on this new frontier.

I stayed on a fifth year at UCLA, 1933–1934, taking a number of courses in Physics, which that year were started at the graduate (Master's degree) level. I hoped that graduate work would also be instituted in the Department of Chemistry. Just before the beginning of the fall semester I went to see Provost Ernest Carroll Moore to urge on him the initiation of graduate work in Chemistry, but he indicated that such a decision must be made at the level of President Robert Gordon Sproul. Therefore, I immediately visited Berkeley, brashly called on him (without an appointment) to press on him the initiation of graduate work in Chemistry in UCLA. His secretary, Miss Agnes Robb, let me go in to see Sproul. He treated me very well but was noncommittal on the graduate work questions. (On this visit I met chemistry professor Wendell Latimer, which may have helped in my admission later to graduate work at Berkeley.)

It soon became apparent that I should instead go on to graduate work at Berkeley. Ramsey urged me to go on to Berkeley, an additional incentive to that furnished by physics professor Adams in his course on Atomic Physics, in which he described the pioneering nuclear research underway at Berkeley.

For my graduate work there could be no place but the University of California at Berkeley. The very name, Berkeley, was magic; it was a distant and almost unattainable mecca. The chemistry staff at Berkeley was legendary, having written the textbooks from which we took our courses at UCLA. There were names such as Joel H. Hildebrand, Wendell M. Latimer, William C. Bray, C. Walter Porter, Gerald E.K. Branch, and, of course, the great Gilbert Newton Lewis, dean of the College of Chemistry. I had become acquainted with his 1923 book *Valence and Structure of Atoms and Molecules*, and was fascinated by it. I wanted to meet and become acquainted with

this remarkable man. The name of the rising young nuclear physicist Ernest O. Lawrence was beginning to ring through the world of science. I wanted to work as near as possible to Lewis (the great "G.N." — "The Chief") and to Lawrence. And again the absence of a tuition fee was consistent with the state of my finances.

Reaching this mecca was not necessarily simple. Not everyone was admitted, and so the custom was to apply to a number of graduate schools. Moreover, I had not only to be accepted but to be granted a teaching assistantship (at a salary of \$50 per month) to support me through graduate study. UCLA chemistry professor James B. Ramsey, who had done his graduate work at Berkeley, assured me that there was no need to apply to alternative institutions, that Berkeley would grant me both my wishes. And so it did, to a lingering disbelief on my part, despite Prof. Ramsey's reassurances.

It is difficult to describe the exciting, glamorous atmosphere that existed at the University of California at Berkeley when I entered as a graduate student in August 1934. I took formal courses in Chemistry from such eminent men as Profs. Axel R. Olson and William F. Giauque, and in Physics from Raymond T. Birge and Robert B. Brode. As a teaching assistant in freshman chemistry my instructor colleagues in the laboratory sections included such men as Joel H. Hildebrand (who always gave the main lectures as well), Wendell M. Latimer, William C. Bray, Giauque and Ermon D. Eastman. Probably the high point of each week was the Tuesday afternoon Research Conference held in Gilman Hall, at which graduate students presented a research paper on a current topic from the literature, which was followed by a faculty member, postdoctoral scientist, or advanced graduate student describing his own recent research. The latter was always in the forefront of scientific research in an interesting area. Here we saw G.N. at his best, sitting at the head of the table which dominated the center of the room, chain-smoking his huge black cigars. He asked questions and stimulated discussion over the whole wide range of Chemistry and Physics in a manner which I have never seen equaled.

Another high point was the weekly evening Nuclear Seminar, covering recent articles from the scientific literature and the current work in the College of Chemistry in the area of Nuclear Science; this seminar was run by Willard F. Libby and Robert D. Fowler, who guided my research until he left. G.N. also always attended these seminars, which added considerably to the excitement. In Le Conte Hall on Monday evenings, there was the Physics Journal Club, presided over by Lawrence, including the brilliant galaxy of J. Robert Oppenheimer, Edwin M. McMillan, Luis W. Alvarez, Philip H. Abelson, Martin D. Kamen, and John J. Livingood, just to mention a few. It was in this atmosphere that I was privileged to carry out my doctoral research in the company of such fellow students as David C. Grahame (who worked with me as my research partner), Kenneth S. Pitzer, Samuel Ruben, and many others.

I made a good start toward realizing my ambition to become a nuclear scientist when I completed my graduate thesis on a nuclear physics project, regarding the inelastic scattering of fast neutrons. After starting this project with Fowler, who moved to Johns Hopkins University, I completed the work with Chemistry Professor George Ernest Gibson. Grahame and I carried out this research in the cavernous auditorium of the abandoned East Hall, an ancient building which had been moved from its original site at the present location of Le Conte Hall to a then vacant spot just to the south of Faculty Glade at about the present location of Morrison Hall. We were forced to perform our experiments during the graveyard shift, because Lewis required the use of the Chemistry Department's sole radium-beryllium source of neutrons during the daytime and evening hours. Our experiments provided what was probably the first unequivocal evidence for the phenomenon of inelastic scattering of fast neutrons. We established a minimum probability (cross section) for this type of reaction in the region of lead and bismuth, an observation that was beyond theoretical understanding at that time but was explained years later as due to the closed nucleon shells of 82 protons and 126 neutrons.

When I obtained my Ph.D. degree in May 1937, I stayed on, continuing my research even though I had no immediate prospect of a job. This was a Depression year and satisfactory positions were very difficult to obtain. Yet such was the atmosphere at Berkeley and my preoccupation with my research that I was only vaguely worried about my future. Then one day in the middle of the summer, G.N. called me in and asked whether I would like to serve as his personal research assistant. Because of his reputation and standing, he was almost unique in having such an assistant, and the position at \$1,800 per year happened to be open at that time. I was overwhelmed at this opportunity and immediately accepted, after first expressing some genuine doubts as to my adequacy. In this role, I published several papers with "The Chief" in the area of generalized acids and bases, which was his current interest and rather far from my own area and aptitude.

Some time before I began my work with G.N. I entered almost by accident the mainstream of my career as a nuclear scientist. One day in 1936 I was suddenly confronted by Jack Livingood, a physicist who was favored by ready access to that nuclear horn of plenty, the 27-inch cyclotron. He literally handed me a "hot" target, just bombarded by the machine, and asked me to process it chemically to identify the radioisotopes that had been produced. Naturally, I jumped at the chance. The facility he offered in Le Conte Hall was hardly luxurious. My best recollection is that it was the custodian's closet and that the resources consisted of tap water, a sink, and a small workbench. With some essential materials bootlegged from the Department of Chemistry, I performed the chemical separation to Jack's satisfaction. In the course of my collaboration with Livingood, covering a period of five years, we discovered a number of radioisotopes which proved useful for biological explorations and medical applications. Among the isotopes that we discovered were iodine-131 and iron-59, and among the useful isotopes that we characterized was cobalt-60.

The discovery of iodine-131 has given me special satisfaction. On one occasion during this period, in 1938, the late Dr. Joseph G.

Hamilton, one of the outstanding nuclear medical pioneers, mentioned to me the limitations on his studies of thyroid metabolism imposed by the short lifetime of the radioactive iodine tracer that was available. He was working with iodine-128, which has a half-life of only 25 minutes. When he inquired about the possibility of finding another iodine isotope with a longer half-life, I asked him what value would be best for his work. He replied, "Oh, about a week." Soon after that, using the 37-inch cyclotron, Jack Livingood and I synthesized and identified iodine-131, with a half-life, luckily enough, of eight days. This isotope is widely used for the diagnosis and treatment of thyroid disease and the diagnosis of other disorders. I have the added satisfaction that my own mother had her life extended by many years as a result of treatment with iodine-131.

Also in 1938, in a collaboration with Emilio Segrè, I was a discoverer of technetium-99m, which has become the most widely used radioisotope for diagnosis in nuclear medicine.

My experience as a radioisotope hunter led eventually to the transuranium elements, a nuclear field that was to become my lifework. My interest in the subject had been aroused soon after I arrived at Berkeley. In the fall of 1934, at the evening Nuclear Seminar presided over by Libby and Fowler, we learned of the experiments by Fermi and his group in Italy. They reported that they had bombarded uranium with neutrons and produced what they thought were radioactive isotopes of transuranium elements, i.e., elements in the periodic table that were heavier than and beyond the heaviest natural element, uranium. Somewhat later, this work was taken up in Germany by Otto Hahn, Lise Meitner, and Fritz Strassmann. I read avidly all the reports on these so-called transuranium elements. I even chose this as my topic for the Tuesday Research Conference, using one of the papers by Hahn and his associates as the basis for a complete description of the chemical properties of these transuranium elements, a nonsubject on which I considered myself already a minor expert.

Then, at the Journal Club meeting in the Department of Physics on a Monday night in January 1939, my mastery of the "field"

vanished in a moment. The information had come through by word of mouth that Hahn and Strassmann in Germany had identified some of the radioactivities as isotopes of barium and lanthanum, and that what actually happened upon the bombardment of uranium with neutrons was the splitting of the uranium nucleus into two approximately equal-sized fragments, with the release of a large amount of nuclear energy. Nuclear scientists had been looking at fission products, not transuranium elements.

I cannot possibly describe either the excitement that this produced in me or the chagrin I felt in realizing that I had failed to interpret correctly the wealth of information I had studied so assiduously for a number of years. After the seminar was over I walked the streets of Berkeley for hours, in turn exhilarated by the beauty of the discovery, despairing over my lack of insight and intrigued by the import of this exciting new fission reaction.



Fig. 4. Helen L. Seaborg and Glenn T. Seaborg, Christmas 1941 in San Francisco.

I have often said that my greatest discovery was Helen Griggs, the girl I married. I first met Helen in September 1938, when she was serving as Ernest Lawrence's secretary. I found her very winsome and felt that I wanted to get to know her better. She is five years younger than I. However, although I saw her many times during the intervening three years, I didn't succeed in dating her until the fall of 1941. I had competition from Donald Cooksey (assistant director of the Radiation Laboratory), who was dating her on a regular basis. Helen and I continued dating during the fall of 1941, and it was clear by Christmas time that I was madly in love with her (Fig. 4). Finally here was a girl that I was dating exclusively. For the first time since I came to Berkeley in 1934, I did not go home to my parents for Christmas, but had Christmas dinner with Helen in San Francisco.



Fig. 5. Helen L. Seaborg and Glenn T. Seaborg at Seaborg's parents' home, South Gate, June 5, 1942, the day before they were married.

When the decision was made that I should move to Chicago for work on the Plutonium Project, I immediately proposed to Helen, on March 23, 1942, and she accepted. The understanding was that I would make a visit back to Berkeley soon to join her for the wedding. After our reunion in Berkeley we visited my parents in South Gate, then boarded a train headed for Chicago, planning to get married en route (Fig. 5). Later Helen wrote the following account of our misadventures in getting to "the altar":

"We had quite a time getting married but it was also very amusing; in fact, we began to feel like a movie scenario before we got through. We got off the train in Caliente, Nevada, on Saturday morning, June 6, about ten o'clock, with a great deal of confidence, without a care in the world, and a feeling that we merely had to take care of a few details and we would then have been quietly married. Little did we realize what was in store for us! We first decided to check our bags, but Caliente had no checkroom. The telegraph operator finally told Glenn that he could leave our things in his place. We deposited our junk and proceeded out to look over the town and find the place to get a marriage license. Being a little coy, we strolled around looking for the place instead of boldly asking anyone. Since we couldn't locate the city hall or anything that remotely resembled same, we finally went in to the town telephone and asked the woman who operated it. She acted as though she had never heard of such a thing as a 'city hall.' We then asked where one could get a marriage license, to which she replied, 'Why, from Evans Edwards' in tones that clearly indicated she thought us terribly stupid not to know that. Glenn asked her where Ev was, and she said, 'Why, down around the corner next to the drug store.' Here her tones indicated that it was the same place it had always been, what was the matter with us anyway. So we proceeded to Ev's to find him leaning back precariously in his swivel chair looking into space. We told him we wanted to get a marriage license. From Ev we learned that one could not

obtain a marriage license in Caliente and that the closest place to get one was Pioche, which was some 25 miles north. Ev was most uncommunicative and didn't seem to have anything to offer in the way of a suggestion for us to get over there. At this point a woman came out. Like most women, she had romance in her soul, and she told us we could probably get a ride in the mail truck, told us where to find it, and said it had not yet left. We thanked her gratefully and left.

"Glenn went to find the driver of the truck while I went to buy a pair of tennis shoes. The town was nothing but dust, and, of course, I had nothing but toeless shoes. When I came back I found him talking to a young man. The latter, it turned out, was a deputy sheriff and son of the telegraph operator, in whose place we had left our bags. Apparently the local inhabitants were somewhat suspicious of us and had asked the young fellow to check up on us. So he had come up to Glenn and asked, 'Are you a teacher?' Poor Glenn, this was a horrible shock; he thought, 'Surely it doesn't stick out all over me already,' but he admitted he was. This was followed with, 'Do you teach Chemistry?' 'Yes.' After this cautious approach the fellow admitted that he had just graduated from Cal majoring in Chemistry [*Glenn learned much later that the deputy's name was Frank White Anders*], that Caliente was his home, and that he was there for the summer before taking a job in research laboratories in Washington this fall. Then he was very nice and offered to help us in any way he could; so he and Glenn went over to the town telephone to call Pioche to make sure we could get in the county courthouse if we got there after noon (since it was Saturday). A voice at the other end of the wire said of course someone would be there — didn't he know the county commissioners were meeting that afternoon? Then we piled into the mail truck and were off, via Panaca, for Pioche. We arrived about 12:40 p.m. to find the assistant county clerk waiting for us. She told us that she would have been gone if Glenn hadn't

called; so we felt very lucky. She made out our license and was more nervous than we were. By that time we were very calm anyway — prepared for whatever might come. I am sure we were beyond the point of surprise. Then she asked us if we wanted her to call the judge, and we asked her about a minister. It turned out there was only one in town; so she tried to locate him for us, but he was nowhere to be found. We decided we had better not take further chances and asked her to get the judge for us then. Well, nobody knew where the judge was; so she told us to get in her car and she would drive us to town to find the minister or the judge. (The courthouse was about a mile from the town itself.) When we got there, she told us to go have lunch while she hunted. We invited her to have lunch with us, but she insisted on our going ahead and that she would hunt. She told us which was the better restaurant in town and went off. When we were half through lunch, she came in to tell us that she had located the judge and to come to her car when we were finished eating. A few minutes later the judge [*Glenn learned later that his name was Edgar L. Nores*] came in and told us they were right across the street waiting for us. I shall never forgive Glenn (as I tell him as frequently as I have an opportunity) for practically telling the judge that we would be there as soon as he finished his apple pie, and he sat there and calmly finished. The rest of the day was comparatively calm after that. The judge was just like the country judge one reads about in books. He was very proud of his country and told us all about the mining there. He was rather cute. When he gave me the marriage license, he told Glenn to keep his hands off — that it was mine — and told me not to let him have it. We got quite a kick out of it. Then we both forgot to pay him — after our having some discussion as to what one should give him.

“Glenn and I were both convinced that had we not the good fortune to run into the two young women with romantic souls we would never have succeeded in getting

married. The assistant clerk was a roly-poly good-natured person who told us that Pioche was not her hometown — Panaca was. (Panaca was 13 miles south.) It turned out she was married to a sailor and had been going where he did whenever she could, but he went to Ireland last summer, and she had felt lonely and had come back to Pioche to work. She had been married eight years and apparently thought it was a very good idea.

"We had to wait until 4:30 for the mail truck to take us back to Caliente; so we took a walk and proceeded up a hill and saw some beautiful cedars and got some sun. We then decided that the city folk may make suckers of the country people who come to town but that this is nothing to what the country folk do to the city guys. We met a man with a bunch of pictures of the Pioche High School band, and it seemed the band had just won some kind of an honor and that they would be able to do something else wonderful if only we bought one of the pictures. He told us that this was the only honor that had ever come to Pioche and put up a wonderful sales talk — what could we do but purchase a picture of the Pioche High School band. On the way back to town we met a very palsy-walsy fellow (slightly tipsy), but he wanted us to stop and have a cigarette with him. We gravely thanked him, told him we were very sorry we didn't smoke. This left him feeling very sad, and he assured us he wasn't mad and that we didn't have to have a cigarette with him. We loafed around and drank cokes until the mail truck started back, and we arrived in Caliente about 5:30. So we say it took us all day to get married. Confidentially, I was beginning to wonder if we were really going to get married, and Glenn doesn't deny that he conjured up these obstacles himself in the hope that he would get out of it at the last moment. The funny part is that we got married in Nevada to save time."

Helen and I visited Caliente and Pioche again at the time of our 50th wedding anniversary, June 1992. We found some changes,

but the Caliente railroad station building was still there, changed to a museum and city offices, and the Lincoln County Courthouse in Pioche was still there, even the room where Justice of the Peace Edgar L. Nores performed our marriage ceremony. Helen and I have succeeded in locating Frank White Anders so we could tender him our belated thanks for his crucial help on June 6, 1942.

My entry into the transuranium field is described in the introductory passages of Chapter 1.

Glossary

Acronyms

ACS	American Chemical Society
ADAM	3 MeV injector for SuperHILAC
ADC	Analog-to-Digital Converter
AEC	Atomic Energy Commission
AFOAT	Armed Forces Office of Atomic Energy
ARCA	Automated Rapid Chemistry Apparatus
BART	Bay Area Rapid Transit
BevaLAC	Combination of Bevatron and SuperHILAC working in tandem, LBL
BGS	Berkeley Gas-filled Separator
CERN	Centre Européenne pour la Recherche Nucléaire, Geneva, Switzerland
CNIC	Committee on the Nomenclature of Inorganic Chemistry
Dees	Accelerating electrodes inside the cyclotron shaped in the letter "D"
DESY	Deutsche Elektronen Synchrotron (German Electron Synchrotron)
DQD	Dipole-Quadrupole-Dipole
DSDV	Dirac-Slater Discrete Variational
ECR	Electron Cyclotron Resonance ion source
ETR	Engineering Test Reactor
FAKE	Fast Automatic Khemistry Experiment
GSI	Gesellschaft für Schwerionenforschung mbH
HADES	Heavy Atom Detection Equipment Studio
HDEHP	di-2-ethylhexylorthophosphoric acid

HEVI	Heavy Element Volatility Instrument
HFIR	High Flux Isotope Reactor
HILAC	Heavy Ion Linear Accelerator
IDCNS	Interdivisional Committee on Nomenclature and Symbols
IUPAC	International Union of Pure and Applied Chemistry
IUPAP	International Union of Pure and Applied Physics
JINR	Joint Institutes for Nuclear Research at Dubna, USSR
KAH	Kernphysikalische Arbeitsgemeinschaft Hessen
LASL	Los Alamos Scientific Laboratory
LANL	Los Alamos National Laboratory (effective 1981)
LBL	Lawrence Berkeley Laboratory
LBNL	Lawrence Berkeley National Laboratory (effective June 16, 1995)
LEAP	Large Einsteinium Accelerator Program
linac	linear accelerator
LN	liquid nitrogen
MGA	Merry-Go-Around, rotating wheel system at LBNL
MIBK	Methylisobutyl ketone
MIT	Massachusetts Institute of Technology
MTR	Materials Testing Reactor
NAO	National Adhering Organizations
NSD	Nuclear Science Division, Lawrence Berkeley National Laboratory
OLGA	On-Line Gas Chemistry Apparatus
ORNL	Oak Ridge National Laboratory
PAC	Pure and Applied Chemistry
RFQ	Radio Frequency Quadrupole
RHIC	Relativistic Heavy Ion Collider
SASSY	Small Angle Separator System
SHE	Super-Heavy Elements
SHIP	Separator for Heavy Ion Reaction Products
SIS	Storage ring Ion Source at GSI
SLAC	Stanford Linear Accelerator Center
SRP	Savannah River Project
SuperHILAC	Improved HILAC (1971–1993)

TBP	Tributylphosphate
TIOA	Triisooctylamine
TOF	Time-of-Flight
TRU	TRansUranium Processing Facility at ORNL
TTA	Thenoyltrifluoracetone
TWG	Transfermium Working Group
UNILAC	UNIversal Linear Accelerator
VW	Vertical Wheel
Z	Atomic number = number of protons
Z > 89	Transactinium Elements
Z > 103	Transactinide Elements

Decay Modes

α	alpha
β	beta
ec	electron capture
SF	Spontaneous Fission

Units

s = second

min = minute

h = hour

d = day

A = ampere

b = barn = 10^{-24} cm²

eV = electron volts

g = gram

m = meter

Prefixes

M = mega = 10^6

k = kilo = 10^3

m = milli = 10^{-3}

micro = μ = 10^{-6}

nano = n = 10^{-9}

pico = p = 10^{-12}

Chapter 1

Introduction

1.1. The Pretransuranium Story

The transuranium story began in Italy soon after the discovery of the neutron in England and of artificial radioactivity in France. In their original work in 1934 in Rome, E. Fermi, E. Amaldi, O. D'Agostino, F. Rasetti, and E. Segrè^{1,1} bombarded uranium with neutrons and obtained a series of β -particle-emitting radioactivities. On the basis of the periodic table of that day (Fig. 1.1), they were led to believe that the first transuranium element, with atomic number 93, should be chemically like rhenium (i.e., eka-rhenium, or Eka-Re), element 94

PERIODIC TABLE - BEFORE WORLD WAR II

1 H																	2 He															
3 Li	4 Be															5 B	6 C	7 N	8 O	9 F	10 Ne											
11 Na	12 Mg															13 Al	14 Si	15 P	16 S	17 Cl	18 Ar											
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr															
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	(43)	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe															
55 Cs	56 Ba	57-71 La- Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	(85)	86 Rn															
(87)	88 Ra	89 Ac	90 Th	91 Pa	92 U	(93)	(94)	(95)	(96)	(97)	(98)	(99)	(100)																			
																		57 La	58 Ce	59 Pr	60 Nd	(61)	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu

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Fig. 1.1. Pre-World War II periodic table. Parentheses indicate elements undiscovered at that time.

like osmium (Eka-Os), and so forth. Therefore, they assigned a 13-min activity to element 93. To quote from the above-mentioned paper entitled "Possible Production of Elements of Atomic Number Higher than 92":

"This negative evidence about the identity of the 13-min activity from a large number of heavy elements suggests the possibility that the atomic number of the element may be greater than 92. If it were element 93, it would be chemically homologous with manganese and rhenium. This hypothesis is supported to some extent also by the observed fact that the 13-min activity is carried down by a precipitate of rhenium sulphide insoluble in hydrochloric acid. However, as several elements are easily precipitated in this form, this evidence cannot be considered very strong."

Soon thereafter, in a paper^{1,2} entitled "*Über das Element 93*" ("On Element 93"), Ida Noddack took issue with this interpretation, suggesting that the radioactivities observed by Fermi *et al.* might be due to elements of medium atomic numbers:

"Es wäre denkbar, dass bei der Beschiessung schwerer Kerne mit Neutronen diese Kerne in mehrere grössere Bruchstücke zerfallen, die zwar Isotope bekannter Elemente, aber nicht Nachbarn der bestrahlten Elemente sind." [One could think that in the bombardment of heavy nuclei with neutrons these nuclei disintegrate into several larger fragments which, although they are isotopes of known elements, are not neighbors of the irradiated elements.]

However, this paper, which intimated the possibility of the nuclear fission reaction, was too revolutionary and was not taken seriously. In addition, experiments in Germany during the following years by O. Hahn, L. Meitner, and F. Strassmann (Fig. 1.2) appeared to confirm the Italian interpretation and for several years the "transuranium elements" were the subject of much experimental work and



Fig. 1.2. Fritz Strassmann, Lise Meitner, and Otto Hahn (left to right), Mainz, Germany, 1956.

discussion. In a typical paper by Hahn, Meitner, and Strassmann,^{1,3} part of a series they published during 1935–1938, they reported a 16-min ${}_{93}\text{Eka-Re}^{237}$, 2.2-min ${}_{93}\text{Eka-Re}^{239}$, 12-h ${}_{94}\text{Eka-Os}^{237}$, 59-min ${}_{94}\text{Eka-Os}^{239}$, 3-day ${}_{95}\text{Eka-Ir}^{239}$, and 12-h ${}_{96}\text{Eka-Pt}^{239}$.

In 1938, I. Curie and P. Savitch,^{1,4} working in Paris, found a product of 3.5-h half-life that seemed to have the chemical properties of a rare earth, but they failed to give a rational interpretation of this astonishing discovery. Their paper had the title “*Sur la Nature du Radioélément de Période 3.5 Heures Formé dans L’Uranium Irradié par les Neutrons*” (“On the Nature of a Radioactive Element with 3.5-Hour Half-Life Produced in the Neutron Irradiation of Uranium”), and included the following:

“Nous avons montré qu’il se forme dans l’uranium irradié par les neutrons un radioélément de période 3.5 heures dont les propriétés chimiques sont semblables a celles des terres rares. Nous la désignerons ci-dessous par la notation $R_{3.5h}$. $R_{3.5h}$ se sépare nettement de Ac, allant en tête, de fractionnement, alors que Ac va en queue. Il semble donc que ce corps ne puisse être qu’un élément transuraniien possédant des propriétés très différentes de celles des autres éléments transuraniens connus, hypothèse qui soulève des difficultés d’interprétation.” [We have shown that

in the neutron irradiation of uranium a radioactive element with a half-life of 3.5 hours is produced, with chemical properties similar to those of rare earths. In the following we will refer to it as $R_{3.5h}$. $R_{3.5h}$ separates cleanly from Ac by going to the 'head' (beginning) of the fractionation while Ac goes to the 'tail' (end). It seems, therefore, that this species cannot but be a transuranic element having properties very different from those of the other known transuranic elements, a hypothesis that raises interpretational difficulties.]

Then came the breakthrough. Early in 1939, Hahn and Strassmann,^{1,5} on the basis of experiments performed in December 1938, and with interpretive help from Meitner, who had been forced to leave Germany, described experiments in which they had observed barium isotopes as the result of bombardment of uranium with neutrons. This historic paper had the title "*Über den Nachweis und das Verhalten der bei der Bestrahlung des Urans mittels Neutronen entstehenden Erdalkalimetalle*" ("On the Identification and the Behavior of Rare Earth Metals Produced in the Neutron Irradiation of Uranium") and contained the following conclusion:

"Als Chemiker müssten wir aus den kurz dargelegten Versuchen das oben gebrachte Schema eigentlich umbenennen und statt Ra, Ac, Th die Symbole Ba, La, Ce einsetzen. Als der Physik in gewisser Weise nahestehende "Kernchemiker" können wir uns zu diesem. allen bisherigen Erfahrungen der Kernphysik widersprechenden, Sprung noch nicht entschliessen. Es könnten doch noch vielleicht eine Reihe seltsamer Zufälle unsere Ergebnisse vorgetäuscht haben." [We, as chemists, based on the briefly described experiments, should rename the above-mentioned scheme and replace Ra, Ac, Th with the symbols Ba, La, Ce. As nuclear chemists, being in some respects close to physics, we have not yet been able to take this leap, which contradicts all previous experiences in nuclear physics. It could be that a series of strange coincidences could have mimicked our results.]

Subsequent work showed that the radioactivities previously ascribed to transuranium elements are actually due to uranium fission products, and hundreds of radioactive fission products of uranium have since been identified. Thus in early 1939 there were again, as five years earlier, no known transuranium elements. During these five years Seaborg developed an increasing interest in the transuranium situation. When as a graduate student he gave his required annual talk at the College of Chemistry's weekly Research Conference in 1936, he chose the transuranium elements as his topic, describing the work of Hahn, Meitner, and Strassmann referred to earlier.

During the two years following Seaborg's seminar talk in 1936 and before the discovery of fission, his interest in the neutron-induced radioactivities in uranium continued unabated and, in fact, increased. He read and reread every article published on the subject. He was puzzled by the situation — both intrigued by the concept of the transuranium interpretation of the experimental results and disturbed by the apparent inconsistencies in their interpretation. He remembers discussing the problem by the hour, with Joseph W. Kennedy, a colleague in research, often in the postmidnight hours of the morning at the old Varsity Coffee Shop on the corner of Telegraph and Bancroft Avenues near the Berkeley campus, where they often went for a cup of coffee and a bite to eat after an evening spent in the laboratory.

Seaborg first learned of the correct interpretation of these experiments, that neutrons split uranium into two large pieces in the fission reaction, at the weekly Monday night seminar on nuclear physics conducted by Ernest O. Lawrence in Le Conte Hall. On this exciting night, January 30, 1939, they heard the news from Germany of Hahn and Strassmann's beautiful chemical experiments. He recalls that at first the fission interpretation was greeted with some skepticism by a number of those present, but, as a chemist with a particular appreciation for Hahn and Strassmann's experiments he felt that this interpretation just had to be accepted. He remembers walking the streets of Berkeley for hours after this seminar in a

combined state of exhilaration in appreciation of the beauty of the work and of disgust at his inability to arrive at this interpretation despite his years of contemplation on the subject.

In investigations by Edwin M. McMillan^{1,6} at Berkeley and others elsewhere, one of the radioactivities produced in the neutron bombardment of uranium was found to behave differently than the others. This beta radioactivity with a half-life of about two days did not separate by recoil from thin layers of uranium, as did the energetic fission products, when uranium was bombarded with slow neutrons. Segrè, thinking that this was an indication that it might not be a fission product, performed a chemical separation to see if it was a transuranium element *produced* as the daughter of the previously well-known β -particle-emitting ^{239}U . He was unsuccessful and published a Letter to the Editor^{1,7} entitled "An Unsuccessful Search for Transuranic Elements," identifying the two-day radioactivity as being due to a rare earth element because of his lack of chemical expertise.

1.2. Early Days at the Berkeley Radiation Laboratory

The invention of the cyclotron at Berkeley by Ernest O. Lawrence and M. Stanley Livingston in 1931^{1,8} started a new era in physics by making a giant leap possible with a relatively simple device. Up to this time progress in the production and study of artificial radioactivities was very slow, because although α -emitting sources prepared from natural ores were used initially in the discovery of artificial radioactivity by the Joliot-Curies, the intensities that could be achieved in this manner were very small. The nonresonant accelerators that soon became available provided much current but could not attain sufficiently high energies to do very much in the way of interesting chemistry or physics. The cyclotron, on the other hand, was a relatively simple device and could be constructed inexpensively by physicists anywhere in the world with only modest help from the Berkeley "cyclotroners."

Lawrence was an amazing person with a tremendous amount of drive. He immediately recognized that his invention would be a boon for research and he soon became a "promoter" and learned how to popularize his creation. This he did by offering the help of his laboratory and its people to other laboratories so that they too could take advantage of this revolutionary new tool. He also became very adept at "scrounging" needed parts from industry at little or no cost to keep the costs down as he continued with his experimentation to build ever-larger cyclotrons. His laboratory started out with a tiny demonstration "proton merry-go-round" that could be held in one hand (without the magnet) and gave an energy of 80 keV for molecular hydrogen ions. After this success new cyclotrons were built with increasingly large diameters of the magnetic pole faces to enable the experimenters to learn more about the details of how the device worked.

The next step was 1.22 MeV and they progressed soon to the 27-inch machine that could make large amounts of radioactivity with 10–15 mA of 6 MeV deuterons. In August 1937 it was converted into the 37-Inch Cyclotron to get several times that current at 8 MeV. As a result of the broad interests of Ernest and his brother John, who was a medical doctor, the scope of investigations in the Radiation Laboratory extended beyond basic research. They included the practical interdisciplinary applications of these new tools of investigation with special emphasis on applications to medicine. Thus one of the chief driving forces to inspire higher currents and higher energies was the need to make large quantities of radiosodium for patient treatment. Later, when Robert Wilson discovered that 90% or more of the beam was lost before extraction, he and Martin Kamen devised probes covered with ferrous phosphide that would skim off this internal wasted beam to make ^{32}P and ^{59}Fe for the biologists; in this way they could meet the needs of biologists and physicists at the same time. In one fell swoop they had multiplied the usefulness of the 37-Inch Cyclotron by an order of magnitude in 1938.

Glenn Seaborg was in the right place at this time to begin his basic contributions to the Laboratory. In an address in 1970 to the

Society of Nuclear Medicine while Chairman of the U.S. Atomic Energy Commission, he told the story of how he became involved with cyclotron research:

"My collaborator in most of this work was a young nuclear physicist, John J. Livingood. Our association extended over a period of about five years (1936–1941) even though he left Berkeley to accept a position at Harvard University near the end of 1938. The transmutation reactions which led to the production of these radioisotopes were effected first with the 27-Inch Cyclotron, which later became the 37-Inch Cyclotron, in the old Radiation Laboratory, and finally with the 60-Inch Cyclotron in the Crocker Laboratory. The old Radiation Laboratory was a picturesque place, crowded almost beyond belief with its large complement of cages full of mice, for use in the experiments by the biomedical pioneers, and apparatus used by the other experimentalists and the cyclotron builders.



Fig. 1.3. Glenn T. Seaborg, Berkeley Campus, April 1936.

"The start of my partnership with Livingood was the result of an essentially accidental encounter. One day in the spring of 1936 (Fig. 1.3) I ran into Jack somewhere in the area between Gilman Hall (the Chemistry Building) and Le Conte Hall (the neighboring Physics Building). (At that time I was a second-year graduate student in the Department of Chemistry working on my thesis problem, which finally emerged as an investigation of the inelastic scattering of fast neutrons.) He told me that he had a target of the element tin which was being bombarded with 5-MeV deuterons in the 27-Inch Cyclotron at that very moment and asked whether I would like to perform the necessary chemical separations in order to make it possible to identify the transmutation products. The chemical operations would have to be performed immediately after the bombardment was completed within the next hour or so.

"Thus I was literally handed a 'hot' target and asked to process it chemically in order to identify the radioisotopes of tin, as well as isotopes of neighboring elements antimony and indium, that might be produced as transmutation products. Naturally I jumped at the chance.

"But the facility he placed at my disposal was hardly luxurious. This turned out to be a corner of a room on the second floor of Le Conte Hall, and the resources consisted of tap water, a sink, a primitive hood, and a small workbench. With some essential materials bootlegged from the Department of Chemistry I performed the chemical separation. Jack's measurements on these chemical fractions showed the presence of some new radioisotopes of tin, an activity in the indium fraction which we later showed to be due to tin (i.e., the indium had been incompletely separated from tin), and two activities in the antimony fraction. We later proved that one of these latter two activities was due to a well-known radioisotope of copper, produced from copper impurity in the tin target, a possibility that I had not anticipated in this, my first, venture into radioisotope hunting.

"Livingood generously included my name as a coauthor in the ensuing publication, entitled 'Deuteron-Induced Radioactivity in Tin,'

which thus became my first scientific publication. It cannot be ranked with my better publications because of the errors in the indium and antimony activities, but perhaps these were excusable on account of the unexpected nature of my encounter with Jack and the necessarily makeshift and hasty chemical separations I had to perform.

"The chemical work I performed during my continuing collaboration with Jack while he was still in Berkeley was all done in this little corner of the room in Le Conte Hall. Our teamwork in every instance consisted of Jack's performing the cyclotron bombardments, after which I dissolved the target material and made the chemical separations. We next mounted my chemical end products on cardboard sheets and covered these with thin cellophane. He then measured their radiation characteristics, using a small Lauritsen quartz fiber electroscope mounted in a sort of cubbyhole room in the basement at the opposite end of Le Conte Hall. Jack constructed and operated the quartz fiber electroscope with consummate skill.

"As our work progressed the measurements all tended to be made on one electroscope which, near the end of 1938, Jack successfully moved to his new home at Belmont, Massachusetts. There he continued measuring the radioactive decay of the many samples he took with him from Berkeley and also measured the decay and absorption properties of additional radioactive samples, which I thereafter mailed to him at Harvard. (The informal process of sending radioactive samples in envelopes by mail was commonly used at that time, a method that has long since been superseded by the much more sophisticated methods required by AEC regulations today.) Jack made the move without missing a single point on our many radioactive decay curves. This indeed became a precious little instrument, critical to our whole research program, simple though it was, and it served this purpose until after the beginning of the war. Then it succumbed when one of his children used it as a plaything. This ended our collaboration, but fortunately most of the key measurements had been made on our samples by this time when the war had called Jack and me to other pursuits.

"My collaboration with Livingood was extracurricular. While it was going on I obtained my Ph.D. degree in May 1937. I then served for two years as the personal research assistant of the great physical chemist and Dean of the College of Chemistry, Gilbert Newton Lewis, and I began, in 1939, my service as a member of the faculty of the University of California, Berkeley. In all instances these occupations were demanding, and had priority on my time and attention.

"Our motivation in searching for new radioisotopes was simply the fascination of exploring an exciting new frontier of science. Usually we gave little thought to the possibility that one of the objects of our search would have practical value. But we were in for some surprises. For one thing, medical researchers on two occasions expressed the hope that radioisotopes of specific elements with desirable half-lives would be found — and we were fortunate enough to hit the jackpot. In addition, as the years passed, we have witnessed the appearance of many a delayed jackpot. At the time of their discovery we would have scoffed at the idea that many radioisotopes would ever have any practical value. Yet some of those that would have been most scorned have turned out to be among the most valuable. And I think there is a lesson here, a lesson that we know well and that everyone should understand: we cannot very often predict the practical applications of basic science, but we can predict that those applications will occur, to the enduring benefit of man.

"Our first immediately practical jackpot came early in the collaboration between me and Jack Livingood. We had learned that Prof. George H. Whipple of the University of Rochester was hoping to have a reasonably long-lived radioisotope of iron for use in his studies of hemoglobin in human blood. A visiting chemist from England, Fred Fairbrother, had begun to collaborate with Livingood in the spring of 1937 on this problem but his visit ended before completion of the work. Livingood turned to me, and this was our second team effort.

"With some advance warning, I was able to plan the chemical separation more carefully, to guard against the mixed results from

the hasty work with tin. Jack bombarded iron with 5.5-MeV deuterons in the 27-Inch Cyclotron. When the target came off the cyclotron, around dinnertime as I remember, my chemical setups were ready.

"In my determination to make an extremely clean chemical separation into fractions containing iron and the expected transmutation products cobalt and manganese, I worked all night making precipitations and repeated reprecipitations. When Jack came to work early in the morning just as the sun was rising, I handed him the three fractions and he began his measurements using his electroscope. The iron fraction was found to contain the radioactive isotope iron-59. When the first production run was made with the cyclotron, the resulting iron-59 was sent by E.O. Lawrence to Dr. Whipple, who performed the first tracer experiments on the blood, and these experiments opened up a new era in our understanding of iron metabolism and blood formation.

"Almost at the same time Dr. John H. Lawrence and his colleagues at Berkeley began studies of the blood, using iron-59, which have continued since at Donner Laboratory at rising levels of sophistication. This work and research by other investigators elsewhere has revolutionized our knowledge of blood formation and dynamics and the management of blood diseases. With the possible exception of iodine, the biological behavior of no other element has been studied so thoroughly. Iron-59 also plays a prominent role today in the diagnosis of various blood dyscrasias.

"Prof. Whipple worked in a group of scientists whose impact on what was then an infant field is still felt. One member of the group was Dr. P.F. Hahn, now deceased, who made significant contributions. Another was Dr. William F. Bale, who is still at the University of Rochester and with whom I still have occasional contact, since he is a member of the AEC's Advisory Committee on Biology and Medicine. Although my current interests no longer bring me in touch with Prof. Whipple, I am told that he still comes to work every day at the University of Rochester — at the age of 92!

"In the course of our investigation we also bombarded cobalt with neutrons and found a very long-lived radioactive isotope of cobalt,

confirming the indications of others that such an isotope existed, and our later work identified this as the now well-known cobalt-60. Jack and I continued this line of investigation by bombarding pure cobalt samples, some with neutrons and some with deuterons furnished by the 27-Inch Cyclotron, followed by very careful chemical separation experiments. These bombardments and chemical separations took place in the summer of 1937, and by the spring of 1938 our decay and radiation absorption measurements had pretty well established the radioactive properties of cobalt-60 for which we eventually measured a half-life of 5.3 years. Today cobalt-60 is estimated to be used annually for over 4,000,000 therapeutic irradiations in the United States alone.

"In the course of subsequent work, and on the basis of our bombardments of iron with deuterons in the 37-Inch Cyclotron, we also discovered and measured the radioactive decay properties of iron-55, eventually shown to have a half-life of about 2.6 years, and cobalt-57, which we later characterized as having a half-life of about 270 days. These latter investigations were not completed until after Livingood left Berkeley in late 1938 to take up his position in the Cyclotron Laboratory at Harvard University. Both of these radioisotopes found their place in the stable of radioisotopes useful in medicine. The soft X-rays emitted by iron-55 have curtailed its clinical applicability. However, it is used in investigative studies with iron-59 when double labeling is necessary. Cobalt-57 is used in the Schilling test, which evaluates the absorption of vitamin B₁₂ from the intestine, particularly in patients suffering from pernicious anemia.

"Perhaps the most interesting of all of my collaborations with Jack Livingood, and one having special personal meaning to me, was the discovery (while he was still in Berkeley) of what is now the work-horse of medical radioisotopes, namely iodine-131.

"One day in the spring of 1938 the late Dr. Joseph G. Hamilton, one of the outstanding nuclear medical pioneers, ran into me on the steps of Le Conte Hall and mentioned the limitations on his studies of thyroid metabolism imposed by the short lifetime of the radioac-

tive iodine that was then available to him. He was working with iodine-128, which has a half-life of only 25 minutes. When he inquired about the possibility of finding an iodine isotope with a longer half-life, I asked him what value would be best for his work. He replied, "Oh, about a week." By this time my partnership with Livingood was in full swing; Jack prepared tellurium targets which we bombarded in the 37-Inch Cyclotron, some with 8 MeV deuterons and some with neutrons, and I put them through chemical separation and identification procedures working in my Le Conte Hall quarters. I recall that we wore gas masks and obstetrical gloves in order to avoid acquiring the dreaded "tellurium breath." We escaped this curse but I don't know whether these precautions were actually necessary.

"We were soon able to identify iodine-131, and luckily enough, its half-life turned out to be eight days. This isotope is now used some two million times a year for the diagnosis and treatment of thyroid disease and in a large number of other applications, including the diagnosis of kidney and liver disorders and function tests of these organs, to screen for pulmonary emboli by lung scans, to locate brain tumors and melanosarcomas and to determine blood or plasma volumes and cardiac output. Furthermore, over 99% of all *in vivo* nuclear medicine procedures utilize this isotope. I have the added satisfaction that my mother had her own life extended by many years as a result of treatment with iodine-131.

"A similar pattern developed in our work on zinc-65, also carried out in 1938 while Livingood was still in Berkeley. Here we followed up on the work of others who had observed a radioisotope of zinc with a half-life of about seven months produced by the deuteron bombardment as well as the proton bombardment of copper. Livingood prepared and bombarded a zinc target with 8 MeV deuterons in the 37-Inch Cyclotron and I performed the chemical separations which made it possible for us during 1938 to assign this radioactivity to zinc-65, characterize its half-life as 250 ± 5 days, and to determine the energy of the radiations, with the help of Jack's measurements with his electroscope. This radioisotope found limited

use in the scanning of the prostate gland and in the study of insulin uptake in the pancreas, but other methods have proved to be more successful.

"My roles in the discovery of technetium-99m and cesium-137 involved collaboration with other colleagues and in these instances we had no premonition concerning their ultimate beneficial applications to medicine when the work was performed. My collaboration with Emilio Segrè began in the summer of 1938 soon after his move, a number of months before his family, from Palermo, Italy, to the United States. On the basis of his familiarity with my publications with Livingood describing our work on new radioactive isotopes, he sought me out as a collaborator. He had participated at Palermo with his colleague C. Perrier in the discovery of the first synthetic element, technetium, with the atomic number 43.

"Segrè was anxious to pursue his investigations of this intriguing new element, and within a few days after our first encounter we had arranged for the bombardment of molybdenum with 8-MeV deuterons in the 37-Inch Cyclotron. In this case we collaborated on the chemical separations, using the techniques worked out by Segrè and his coworkers at Palermo, and we isolated a fraction designed to contain any isotopes of the element with the atomic number 43. We performed our chemical separations in a rather well-equipped little chemistry laboratory situated in the newly occupied Crocker Laboratory. We measured the radioactive decay and the radiation absorption properties through the use of an ionization chamber connected to a vacuum tube electrometer system — a rather more sophisticated detection apparatus than that which Livingood and I had been employing. To our delight, we discovered an isotope of great scientific interest, because it decayed by means of an isomeric transition with emission of a line spectrum of electrons coming from an almost completely internally converted gamma ray transition.

"This concept was so new to the experts in the Radiation Laboratory, including J.R. Oppenheimer and E.O. Lawrence, that we were asked to delay our publication until additional measurements and interpretations could be evolved. In the meantime, Segrè's former

Italian colleague, Bruno Pontecorvo, then working in Paris, observed and published a description of a similar isomeric transition which he had observed in an isotope of rhodium. This was a form of radioactive decay which had never been observed before this time.

"Segrè and I were able to show that this radioactive isotope of the element with the atomic number 43 (later given the name 'technetium' by Perrier and Segrè) decayed with a half-life of 6.6 h and that it was the daughter of a 67-h molybdenum parent radioactivity. This chain of decay was later shown to have the mass number 99, and after the convention of designating isomeric states of measurable half-life with the symbol 'm,' meaning 'metastable,' was established, the 6.6-h activity acquired the designation 'technetium-99m.'

"Its radiation quality, short half-life and pharmacologic versatility make technetium-99m ideally suited for medical use. The physical characteristics allow the physician to administer high doses at a substantially lower irradiation exposure to the patient and, because of the higher count rates, greater sensitivity in sequential imaging. There are now molybdenum-99 generators of technetium-99m in hospitals all over the United States and in many other parts of the world. This radioisotope is used in more than 2,000 diagnostic procedures daily in the United States alone involving the diagnosis of thyroid, liver, brain and kidney disorders. (Note: Today, 1998, technetium-99m is the workhorse of nuclear medicine, used in some 10,000,000 diagnostic procedures per year.)

"My involvement with the discovery of cesium-137 presents an entirely different story. After the discovery of fission by Hahn and Strassmann in December 1938 many radioactive fission products were characterized in laboratories all around the world, but the evidence concerning radioactive isotopes of rubidium and cesium was scanty or lacking. Therefore, in the late fall of 1940 I asked an undergraduate student, Margaret Melhase (now Mrs. Robert Fuchs), to take some uranium which had been bombarded with neutrons furnished by the 60-Inch Cyclotron and make chemical separations designed to look for hitherto unknown radioactive fission products such as cesium. She performed her chemical separations on the top

floor of the old 'Rat House,' which even at that time was an ancient ramshackle wooden building dating from the earliest days of the Department of Chemistry at Berkeley. Her measurements of the radioactive decay and radiation absorption properties were performed through the use of a Lauritsen quartz fiber electroscope situated in the cavernous auditorium of the abandoned East Hall. This was an ancient building which had been moved from its original site at the present location of Le Conte Hall to a spot just to the south of Faculty Glade at about the present location of Morrison Hall on the Berkeley campus. (These, by the way, were the quarters in which, working the graveyard shift during the winter of 1936-37, I had performed my Ph.D. thesis experiments on the inelastic scattering of fast neutrons.)

"Miss Melhase also used a Lauritsen electroscope situated on the third floor of Gilman Hall in the latter stages of her research after she had graduated to this higher status. She continued her work until the summer of 1941, by which time she had established the presence of a very long-lived radioactive fission product in the cesium fraction which on the basis of subsequent work we can now identify as being due to the 30 year cesium-137. This isotope has found substantial applications as a gamma ray source in medical therapy similar in its use to that of cobalt-60.

"This, then, has been a brief account of the manner in which our early work at Berkeley led to a number of practical, and largely unanticipated, benefits to mankind.

"In closing, let me say that, despite your remarkable achievements in the last three decades, I am confident that you are only at the beginning of the application of radioisotopes for human welfare. No one could have imagined, back in the 1930s, that you could have accomplished as much as you have. Nor can we visualize the uses of radioisotopes by the close of this century. That these tools will be of immeasurable benefit in the prevention and alleviation of human suffering, and in ways we cannot now imagine, seems assured."

But Lawrence was not content. As far as he was concerned more energy was the name of the game — and his next jump was to a

cyclotron with a pole tip of 60 inches! When asked why it was so big he replied, "Because we can get the money for it!" But the problems do not increase linearly with the diameter of the pole tips, they increase as some much higher power, and although Lawrence had raised the money to build it he also needed first class engineering help. He was fortunate, in that William Brobeck turned up to work for nothing. This was the man who took over the complicated tasks of building Lawrence's dreams, the 60-Inch and the 184-Inch Cyclotrons. In February 1939 beam hunting began in earnest, but it was not until September that operation of the 60-Inch Cyclotron could be called "routine" when a system of crews was set up so that the most efficient use of the time could be made. Figure 1.4 shows Lawrence and some of his people in front of the very large magnet before the vacuum chamber was installed. It was Brobeck who organized the builders of the 184-Inch Cyclotron on an industrial, almost military scale, setting up a "directing committee" as supervisors; even safety, especially electrical, was given a prominent place. The bureaucracy was needed to get the jobs done on time; prior to this time cyclotrons had been constructed in a somewhat haphazard way. This was the

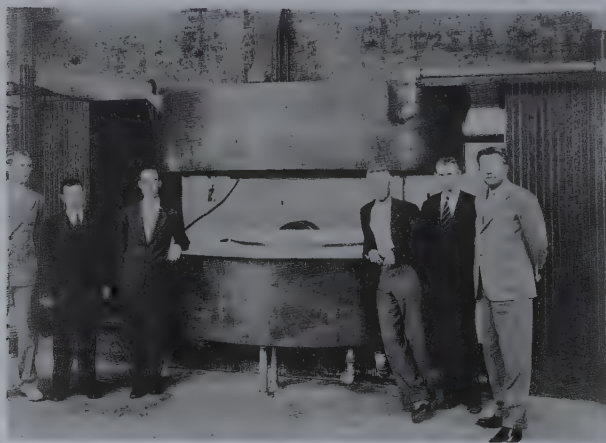


Fig. 1.4. The Crocker Laboratory 60-Inch Cyclotron magnet. *Left to right:* Luis Alvarez, Bob Serber, Bill Brobeck, Donald Cooksey, Edwin McMillan, Ernest O. Lawrence.



Fig. 1.5. Ernest O. Lawrence, Glenn T. Seaborg, and J. Robert Oppenheimer, in early 1946 at the controls to the magnet of the 184-Inch Cyclotron, which was being converted from its wartime use to its original purpose as a cyclotron.

first example of the subordination of the individual to the group; big time science had now arrived and construction of the needed large magnet proceeded based on a scale model made by the young Luis Alvarez. The latest ideas on the large vacuum chamber and the “Dees” were incorporated in the design of the huge machine and work proceeded day and night at a fever pitch. The 184-Inch would be the world’s largest cyclotron for quite some time and its magnet would be extremely important almost immediately for the separation of ^{235}U for the Manhattan atomic energy project (Fig. 1.5).

The 60-Inch Cyclotron was used to make the first two transuranium elements, neptunium (93) and plutonium (94), in 1940 and 1941, and macro amounts of plutonium in 1942 so that Seaborg’s chemists at the Metallurgical Laboratory at the University of Chicago could design and test the chemical process that was to be used at Hanford when the neutron reactors came on-line in 1944 (see Chapter 3). An intense current of deuterons bombarded a beryllium target around the clock, day after day, for months to make the neutrons that were captured in large cans of uranyl nitrate stacked around the cyclotron target. The “hot” material was then sent to the Met Lab, where it was

worked up by a team of chemists. When purified, it resulted in the first plutonium to be seen without the benefit of a microscope. The cyclotron at Washington University in St. Louis also contributed to the early production of microscopic quantities of plutonium.

1.3. Transplutonium Elements

While the Seaborg group was still in Chicago they started their first efforts to make transplutonium elements by sending targets of ^{239}Pu to Berkeley for bombardment with ^4He ions in the 60-Inch Cyclotron. This resulted in the discovery of element 96 in the form of $^{242}\text{96}$, identified in 1944 by a distinctive alpha energy and half-life (see Chapter 4). When the bombardment was repeated months later they were puzzled when they observed a different isotope, $^{240}\text{96}$, with a higher alpha energy and a shorter half-life. The mystery was solved when they found that the cyclotron energy had been increased substantially by another modification in between the two bombardments; they deduced that the excitation functions must be much narrower in width than they had expected. By interposing energy absorbing foils over the target they were able to take advantage of this new tool to fractionate the isotopes and this became a routine procedure in future research. The long distance bombardments continued until the group transferred to Berkeley in 1945–1946.

Now the shorter half-lives became available for investigation, and after the protracted period that it took for the group to set up in its new quarters, elements 97 and 98 were discovered in 1949–1950, as discussed in Chapter 5. The chief reason for the delay was that there were great hazards in dealing safely with the very radioactive americium and curium targets before, during, and after the bombardments and this demanded a lot of special attention. A new one-story laboratory, Bldg. 5, was built for this purpose and there the famed Berkeley glove boxes were developed by Nels Garden and his group. In addition, new methods of detecting α - and X-radiation had to be worked out to detect the meager amount of activity after

it had been separated from the target. All of this preparation was put to good use in 1952 in the work done on the Mike bomb debris (see Chapter 6).

By 1955, the group had progressed to the point where it was ready for what might be called the *tour de force* of all the research on transuranium elements. For the first time, the discovery of a new element was made one atom at a time by means of a new technique. This was element 101, mendelevium, and its discovery is described in detail in Chapter 7. To make the experiment feasible it was necessary to increase the beam density in the 60-Inch Cyclotron by an order of magnitude over its normal value. Bernard Rossi, the cyclotron supervisor, worked hard at this and succeeded by taking advantage of the fringe magnetic field, but it meant that a special probe had to be designed for the experiment. This was the first recoil experiment to be performed for this type of nuclear chemistry/physics and its success led to its use for all of the following discovery experiments.

The Berkeley group had gotten this far by using neutron capture reactions in nuclear reactors or from helium ion (α -particle) bombardments in the Cyclotron. To go higher in atomic number with the target materials that were then available, it was clear that ions heavier than helium were needed. A decade earlier, Alvarez had demonstrated that fully stripped carbon ions could be accelerated in the big cyclotron but at intensities many orders of magnitude lower than would be needed, so Ghiorso and Rossi undertook a regular program at the 60-Inch Cyclotron to try to improve the output. After some months of research, they discovered that the C^{6+} ions were not produced at the ion source; they were produced by a two-step process. They theorized that carbon ions with two electrons missing were produced at the ion source and accelerated at one third the normal velocity with a good intensity. These ions occasionally would collide with gas molecules and be fully stripped and then be accelerated in the normal way. This third harmonic type of operation was observed for other heavy particles also. It led to a poor energy

distribution but it was better than nothing, and the group was able to do some important preliminary experiments and point up the need for a dedicated heavy ion accelerator. Alvarez, fresh from his success with his proton linear accelerator that had just been completed at Berkeley, proposed that a linear accelerator be used for the heavy ions with the difference that each drift tube would contain a magnetic strong focusing element. A joint study/design group was set up with the Yale Physics Department for such a machine and after many months they came up with a Heavy Ion Linear Accelerator (HILAC) along the lines that Alvarez had suggested.

At this stage an amusing incident caused by Lawrence himself occurred that could have changed the history of heavy ion acceleration at Berkeley. On a Sunday in 1955 the Lab had its first Open House celebration and during the event Lawrence sought out Ghiorso, who happened to be there. He asked how the HILAC design was going and when told that it had been finalized and construction was about to begin, he said that he had been thinking that maybe the possibility of building a cyclotron instead of a linac should be reconsidered! He suggested that a meeting of all of the Lab experts should be convened the next day to make sure that the correct decision had been made. One didn't easily dissuade Lawrence in those days, so the meeting was held and in a small group Ernest could be a dominating personality. He had a lot of good points to make, but his suggestion at that time would have delayed the project several more years, because to build a machine equivalent to the HILAC would have meant a very large cyclotron. Ghiorso knew this and searched for the powerful counterargument that would save the linac. He found it when he pointed out that extraction of the beam was a problem for cyclotrons at that time, a figure of 10% being optimistic, whereas extraction from a linac was automatically 100%. That argument, reiterated over and over, finally carried the day. In retrospect, that was a very important decision, because the inventions of the Omnitron and the BevaLAC that followed the HILAC would not have occurred and this in turn would have meant that RHIC would not have been built.

The AEC authorized the construction of the twin accelerators at Berkeley and Yale and they were promptly built. The HILAC consisted of a 750 kV Cockcroft-Walton injector that produced ions with a low charge state for injection into a 30-foot-long, grid-focussed prestripper linac to accelerate the heavy ions up to 1.0 MeV/A. These ions were then passed through a cloud of mercury vapor to strip more electrons from each ion to raise it to a higher charge state and thus minimize the necessary length of the second tank. The second tank was a 90-foot-long linac with magnetically strong focussed drift tubes that took the ions up to 10 MeV/A. The final energy was fixed and the pulse rate was only 2 Hz. The tanks were 10 feet in diameter, so there was a lot of internal surface area to be outgassed. It turned out that the low duty cycle (a pulse rate of 2 Hz with a pulse width of only 2 ms) meant that it was difficult to inject enough power to break through the various "ion locks" very quickly, and as a consequence it was many months before the vacuum was good enough to allow high RF voltage gradients to be established so that acceleration experiments could begin.

1.4. Current Status

The first real transuranium element (neptunium, 93) was identified in 1940. Since then, we have seen the synthesis and identification, i.e., the discoveries, of 20 elements with atomic numbers greater than that of uranium (element 92) — the transuranium elements. These discoveries have added more than 20% to our list of chemical elements and the study of these elements has added much to our understanding of nuclear and atomic structure, the periodic table, nuclear fission and the limits to nuclear stability, and nuclear reaction mechanisms.

The current periodic table, as of April 1998, is shown in Fig. 1.6. The elements through 112 are now known. The names and symbols we shall use throughout this book are those approved by The International Union of Pure and Applied Chemistry (IUPAC) in August 1997 or earlier, except for element 105, for which we shall

continue to use hahnium (Ha), in honor of the great German scientist and codiscoverer of fission, Otto Hahn. Table 13.5 lists the names and symbols approved by IUPAC in August 1997, and Chapter 13 gives a detailed discussion of the many controversies concerning priority of discovery and the naming of the transfermium elements. A time-line for the discovery of the transuranium elements is shown in Fig. 1.7. Strangely enough, the production of Elements 99 (einsteinium) and 100 (fermium) was not planned; they were produced as "by-products" of the first thermonuclear device, tested by the USA in the South Pacific in November 1952. This remarkable story is recounted in Chapter 6. The elements heavier than fermium (100) cannot be produced at reactors via neutron capture and must be produced via light or heavy ion bombardments at suitable accelerators.

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Fig. 1.6. Periodic table as of April 1998.

Discovery of Transuranium Elements

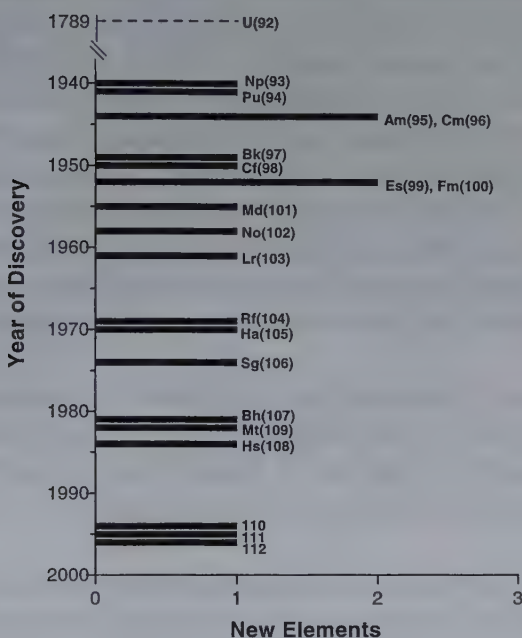


Fig. 1.7. Time-line for discovery of the transuranium elements.

All of the transuranium elements through 101 (mendelevium) were first discovered using chemical separation techniques. However, mendelevium was the first element to be produced and chemically identified on an “atom at a time” basis at an accelerator; its discovery is described in Chapter 7. One-atom-at-a-time chemistry has now been conducted on the elements through seaborgium (106) and plans are underway to extend these studies still further.

Beyond mendelevium the elements were first identified via detection of their nuclear decay and new techniques had to be devised for their positive identification. This is one reason for the ensuing controversies concerning the discoveries of element 102 and heavier. The controversies over priority of discovery and naming are discussed in detail in the chapters on those elements (Chapters 8

through 12), as well as in Chapter 13. The elements from 102 through 106 (seaborgium) were first produced using "hot fusion" reactions in which the initially produced compound nucleus is highly excited and a competition ensues between neutron emission and prompt fission before the desired product can be reached. The ever-increasing probability for prompt fission leads to lower and lower cross sections, and beginning with element 107 the "cold fusion" reaction using targets near the doubly magic ^{208}Pb region with heavy projectiles to produce a less excited compound nucleus has been utilized. A discussion of these reactions and the advances in instrumentation required to extend the known elements to 112 is given in Chapters 11 and 12.

Some reported discoveries of superheavy elements (SHE), how scientists can sometimes be led astray, and current plans to produce them are reviewed in Chapter 14. Finally, in Chapter 15, we reflect on the past and try to use our "cloudy crystal ball" to predict the future.

Many of the transuranium elements can be produced and isolated in large quantities through the use of neutrons furnished by nuclear fission reactors. Plutonium can be produced by the ton; neptunium, americium, and curium by the kilogram; berkelium by the 100 milligrams; californium by the gram; einsteinium by the milligram, and fermium in only sub-picogram quantities. Transuranium isotopes have found many practical applications — as nuclear fuel for the large scale generation of electricity; as compact, long-lived power and heat sources for use in space exploration; as means of diagnosis and treatment in nuclear medicine; in pacemakers and smoke detectors; as tools in numerous industrial processes; in agriculture; and in research in the arts and humanities.

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Chapter 2

Neptunium and Plutonium

2.1. Discovery and Isolation of Neptunium

Toward the spring of 1940, McMillan (Fig. 2.1) began to come to the conclusion that Segrè's conclusion must be wrong^{1,7} that the 2-d β -emitter found in the products of neutron irradiations of uranium was due to a rare-earth element, and that the 2.3-d activity produced in the bombardment of uranium with neutrons might actually be due to the daughter of the 23-minute uranium-239 and thus might,



Fig. 2.1. Edwin M. McMillan, Berkeley, June 8, 1940.



Fig. 2.2. Philip H. Abelson, 1940.

indeed, be an isotope of element 93 with the mass number 239 (93-239). Philip H. Abelson (Fig. 2.2) joined him in this work in the spring of 1940, and together they were able to separate chemically and to identify and thus discover^{2.1} element 93. They showed that element 93 has chemical properties similar to those of uranium, and not similar to those of rhenium as suggested by the periodic table of that time (Fig. 1.1). Element 93 was given the name neptunium (symbol Np) after the planet Neptune, because this planet is beyond the planet Uranus, for which uranium was named.

The early chemistry of neptunium, like that of plutonium (for which ^{238}Pu was used), was limited to studies by the tracer technique using the short-lived isotope, ^{239}Np . Fortunately, there is another isotope of neptunium, ^{237}Np , which was discovered early in 1942 by A.C. Wahl and Seaborg at the University of California,^{2.2} and which is sufficiently long-lived to make work with weighable amounts possible. This isotope is the decay product of the previously known seven-day, β -particle emitter, ^{237}U , which is formed as the result of

an ($n, 2n$) reaction on ^{238}U , and is an α -emitter of relatively long half-life, 2.20×10^6 years.

The first weighable amounts of ^{237}Np were produced by the bombardment of large amounts of uranium with the fast neutrons from the cyclotron. The reaction for the production of this isotope is such that the yield is considerably less than that for ^{239}Pu , and therefore the total amounts available for the early work on neptunium were even smaller than the tiny amounts of this isotope (see below). Nevertheless, L.B. Magnusson and T.J. LaChapelle, working with J.C. Hindman at the Metallurgical Laboratory of the University of Chicago, were able, during 1944, to isolate^{2,3} the element in the form of pure compounds and to study a number of its important chemical properties. The equipment and techniques were the same as those used for the study of plutonium (see below). A picture of the first neptunium, 10 μg of the oxide (NpO_2), isolated in July 1944, is shown in Fig. 2.3. Larger amounts of this isotope were later available. It

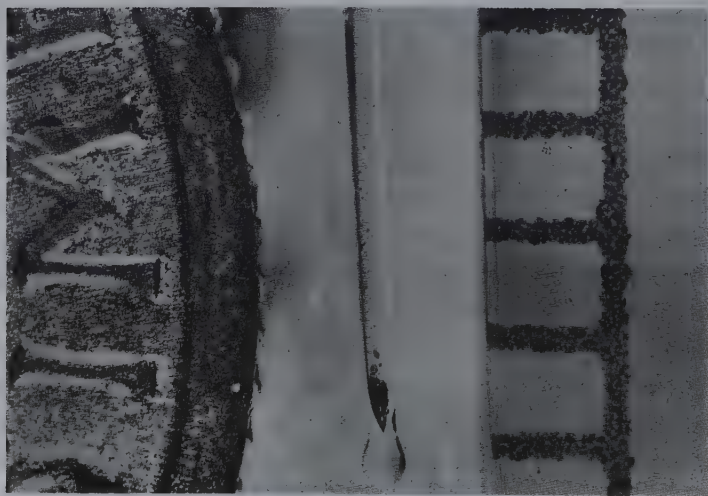


Fig. 2.3. First neptunium compound [10 μg of oxide at bottom of capillary tube (magnified about 20-fold) isolated in July 1944 by L.B. Magnusson and T.J. LaChapelle]. To the right of the sample, for purposes of comparison, is a mm scale, and to the left of the sample a U.S. dime.

is very fortunate, indeed, that the large chain-reacting units produce this isotope, and that by means of special chemical extraction procedures it has been possible to extract and make it available for chemical studies. Using such material, it has been possible to make an intensive study of the chemical properties of neptunium, leading to a thorough understanding of its chemical properties. S. Fried and N.R. Davidson, at the wartime Metallurgical Laboratory, produced the first pure neptunium halides, and they also produced the first neptunium metal by the reduction of neptunium trifluoride with barium.^{2,4}

2.2. Discovery of Plutonium

During the summer and fall of 1940, McMillan started looking for the daughter product of the 2.3-d activity, which obviously would be the isotope of element 94 with mass number 239 (94-239). Not finding anything he could positively identify as such, he began to bombard uranium with deuterons in the 60-Inch Cyclotron in the hope that he might find a shorter-lived isotope — one of a higher intensity of radioactivity that would be easier to identify as an isotope of element 94. Before he could finish this project, he was called away to work on radar at the Massachusetts Institute of Technology (MIT).

During this time Seaborg's interest in the transuranium elements continued. Since he and McMillan lived only a few rooms apart in the Faculty Club, they saw each other quite often and much of their conversation, whether in the laboratory, at meals, in the hallway, or even going in and out of the shower, had something to do with element 93 and the search for element 94. McMillan's sudden departure for MIT in November came as something of a surprise to Seaborg — especially since he did not even know when McMillan had left.

In the meantime, Seaborg had asked Arthur C. Wahl (Fig. 2.4), one of his two graduate students, to begin studying the tracer chemical properties of neptunium with the idea that this might be a

good subject for his thesis. Seaborg's other coworker was Joseph W. Kennedy (Fig. 2.5), a fellow instructor at the University who was also very interested in the general transuranium problem.

When Seaborg learned that McMillan had gone, he wrote to him asking whether it might not be a good idea if he carried on the work he (McMillan) had started, especially the deuteron bombardment of uranium. McMillan readily assented.



Fig. 2.4. Arthur C. Wahl, c. 1946, in his office/laboratory at Washington University, St. Louis, Missouri.

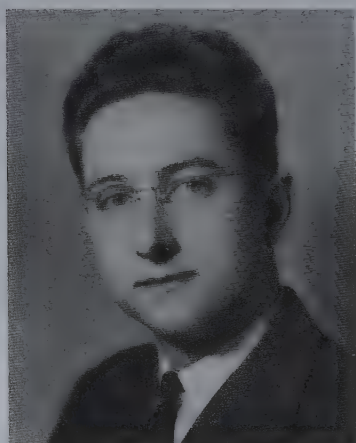


Fig. 2.5. Portrait of Joseph W. Kennedy, 1941.

His first deuteron bombardment of uranium was conducted on December 14, 1940. What was bombarded was a form of uranium oxide, U_3O_8 , which was literally plastered onto a copper backing plate. From this bombarded material Wahl isolated a chemical fraction of neptunium. The radioactivity of this fraction was measured and studied using instrumentation constructed by Kennedy. They observed that it had characteristics different from the radiation from a sample of pure neptunium-239. The β -particles, which in this case were due to a mixture of neptunium-239 and the new isotope of neptunium with mass number 238, had a somewhat higher energy than the radiation from pure neptunium-239 and there was more gamma radiation. But the composite half-life was about the same, namely two days. However, the sample also differed in another very important way from a sample of pure neptunium-239. Into this sample there grew an α -particle-emitting radioactivity. A proportional counter was used to count the α -particles to the exclusion of the β -particles. This work led them to the conclusion that they had a daughter of the new isotope neptunium-238 — a daughter with a half-life of about 50 years and with the atomic number 94. This is much shorter-lived than the now known half-life of $^{239}94$, which is about 24,000 years. The shorter half-life means a higher intensity of α -particle emission, which explains why it was so much easier to identify what proved to be the isotope of element 94 with the mass number 238 (94-238). (Later it was proved that the true half-life of what they had, i.e., 94-238, is about 90 years.)

On January 28, 1941, they sent a short note to Washington describing their initial studies on element 94; this communication also served for later publication in *The Physical Review* in 1946 under the names of Seaborg, McMillan, Kennedy, and Wahl.^{2,5} They did not consider, however, that they had sufficient proof at that time to say they had discovered a new element and felt that they had to have chemical proof to be positive. So, during the rest of January and into February, they attempted to identify this alpha activity chemically.

Their attempts proved unsuccessful for some time. They did not find it possible to oxidize the isotope responsible for this alpha

radioactivity. Then they asked Wendell Latimer to suggest the strongest oxidizing agent he knew for use in aqueous solution. At his suggestion they used peroxydisulfate with argentic ion as catalyst.

On the stormy night of February 23, 1941, in an experiment that ran well into the next morning, Wahl performed the oxidation that gave them proof that what they had made was chemically different from all other known elements. That experiment, and hence the first chemical identification of element 94, took place in Room 307 of Gilman Hall, the room that was dedicated as a National Historic Landmark, 25 years later. Thus, they showed that the chemical properties of element 94 were similar to those of uranium and not like osmium (as suggested by Fig. 1.1).

The communication to Washington describing this oxidation experiment, which was critical to the discovery of element 94, was sent on March 7, 1941, and this served for later publication in *The Physical Review* in 1946 under the authorship of Seaborg, Wahl, and Kennedy.^{2,6}

How element 94 eventually got the name “plutonium” is an interesting story and one worth telling. This work was carried out under self-imposed secrecy in view of its potential implications for national security. Following the discovery in February 1941 and well into 1942, they used only the name “element 94” among themselves and the few other people who knew of the element’s existence. But they needed a code name to be used when they might be overheard. Someone suggested “silver” as a code name for element 93, and they decided to use “copper” for element 94. This worked fine until, for some reason, it became necessary to use real copper in their work. Since they continued to call element 94 “copper” on occasion, they had to refer to the real thing as “honest-to-God copper.”

Going on to more serious possibilities, they considered the names “extremium” and “ultimum” — because they thought they had reached the limit of the periodic table!

The first time a true name for element 94 seemed necessary was in writing the report to the Uranium Committee in Washington in March 1942, which was published in 1948 under the authorship of

Seaborg and Wahl.^{2,7} There were debates within the small group as to what the name should be. It eventually became obvious that they should follow the lead of McMillan, who had named element 93 “neptunium” because Neptune is the next planet after Uranus, which had served as the basis for the naming of uranium 150 years earlier. Thus they should name element 94 for Pluto, the next planet beyond Neptune. But, and this is a little-known story, it seemed that one way of using the base name “Pluto” was to name the element “plutium.” They debated the question of whether the name should be “plutium” or “plutonium,” the sound of which they liked much better. They finally decided to take the name that sounded better. A wise choice, because it is also etymologically correct.

There was also the matter of the need for a symbol. Here, too, a great deal of debate was engendered because, although the symbol



Fig. 2.6. Glenn T. Seaborg and astronomer Clyde W. Tombaugh, discoverer of the planet Pluto, at a press conference, June 10, 1991, at the Sandia National Laboratories on the occasion of a colloquium Seaborg gave about the 50-year history of the discovery of the transuranium elements. Seaborg (age 79) and Tombaugh (age 85) spent time reminiscing with the news media about their discoveries. Seaborg is wearing his “periodic table” necktie and Tombaugh is wearing his “Walt Disney’s Pluto” wrist-watch, Albuquerque, New Mexico. (Courtesy of Sandia National Laboratories.)

might have been "Pl," they liked the sound of "Pu" — for the reason one might suspect. They decided on "Pu," and they expected a much greater reaction after it was declassified than they ever received.

Seaborg had the pleasure of meeting for the first time Clyde Tombaugh, the discoverer of the planet Pluto, in Albuquerque, New Mexico, on June 9, 1991 (Fig. 2.6). At that time, Tombaugh told Seaborg he had also considered naming his planet after the Greek god Cronus or Roman goddess Minerva (rather than after Pluto). In that case, the discoverers would likely have given element 94 the name "cronium" or "minervium," and therefore people throughout the world would never have heard the word "plutonium," which is so much in the news today.

2.2.1. Nuclear Fission of Plutonium

Almost concurrent with this work was the search for, and the demonstration of the fission of, the isotope of major importance — plutonium-239, the radioactive daughter of neptunium-239. Emilio Segrè (again hampered by his lack of chemical expertise) participated in this work together with Kennedy, Wahl, and Seaborg. The importance of plutonium stems from its fission properties and its capability of production in large quantities. This work involved the 60-Inch Cyclotron, the Old Chemistry Building, the Crocker Laboratory, and the 37-Inch Cyclotron, all of which have by now been removed from the Berkeley campus. The 0.5- μ g sample on which the fission of ^{239}Pu was first demonstrated was produced by transmutation of uranium with neutrons from the 60-Inch Cyclotron; it was chemically isolated in rooms in the Old Chemistry Building and the Crocker Laboratory and in Room 307 Gilman; and the fission counting was done using the neutrons from the 37-Inch Cyclotron.

A sample of uranyl nitrate weighing 1.2 kg was distributed in a large paraffin block (neutron-slowng material) placed directly behind the beryllium target of the 60-Inch Cyclotron and was bombarded for two days with neutrons produced by the impact of the full deuteron beam on beryllium. The irradiated uranyl nitrate

was placed in a continuously operating glass extraction apparatus, and the uranyl nitrate was extracted into diethyl ether. Neptunium-239 was isolated from the aqueous layer by use of the oxidation-reduction principle (described later in the next chapter) with lanthanum and cerium fluoride carrier and was reprecipitated six times in order to remove all uranium impurity. Measurement of the radiation from the neptunium-239 made it possible to calculate that 0.5 μg was present to yield plutonium-239 upon decay. The resulting α -activity corresponded to a half-life of 30,000 years for the daughter plutonium-239, in demonstrable agreement with the present best value for the half-life of 24,360 years.

The group first demonstrated, on March 28, 1941, with the sample containing 0.5 μg of plutonium-239, that this isotope undergoes slow neutron-induced fission with a probability of reaction comparable to that of uranium-235. The sample was placed near the screened window of an ionization chamber that could detect the fissions of plutonium-239. Neutrons were then produced near the sample by bombarding a beryllium target with deuterons in the 37-Inch Cyclotron of Berkeley's "Old Radiation Laboratory" (the name applied to the original wooden building, since torn down to make way for modern buildings). Paraffin around the sample slowed the neutrons down so they would be captured more readily by the plutonium. This experiment gave a small but detectable fission rate when a 6- μA beam of deuterons was used. To increase the accuracy of the measurement of the fission cross section, this sample, which had about 5 mg of rare-earth carrier materials, was subjected to an oxidation-reduction chemical procedure that reduced the amount of the carrier to a few tenths of a milligram. A fission cross section for plutonium-239 some 50% greater than that for uranium-235 was found, agreeing remarkably well with the accurate values that were determined later. This result was communicated to Washington on May 29, 1941, and this served as the basis for the later publication of a declassified, expurgated version in 1946 by Kennedy, Seaborg, Segrè, and Wahl.^{2,8}

The observation that plutonium-239 is fissionable with slow neutrons provided the information that formed the basis for the U.S. wartime Plutonium Project of the Manhattan Engineer District (MED) centered at the Metallurgical Laboratory of the University of Chicago. Given impetus by the entry of the U.S. into the war in December 1941, Seaborg and some of his colleagues moved to Chicago in the spring of 1942. The mission of the Met Lab was to develop (1) a method for the production of plutonium in quantity and (2) a method for its chemical separation on a large scale.

2.2.2. Epilogue: ^{244}Pu in Nature

Subsequent to the discovery of ^{244}Pu (see Chapter 6) in the Mike debris, Paul Kuroda in 1960 postulated^{2,9} the existence of ^{244}Pu in the early solar system. In 1971, Alexander *et al.*^{2,10} measured the ratios of xenon isotopes from its spontaneous fission (SF) and found they agreed with those found in achondritic meteorites, thus strongly supporting this hypothesis and the SF decay of ^{244}Pu . Several attempts were then made to detect residual ^{244}Pu in nature, but their sensitivity was too low, i.e., it was well above the upper limit for the average terrestrial abundance of 3×10^{-25} g/g, which can be calculated by assuming that 4.7×10^9 years ago (the approximate age of our solar system), its abundance was equal to the present average terrestrial abundance of ^{232}Th (4.4×10^{-8} g/g), the ultimate decay product of ^{244}Pu . (The α to SF branching ratio for ^{244}Pu is ~ 830 .)

Darleane Hoffman and her coworkers at Los Alamos decided to try to find an ore in which plutonium might be highly enriched in order to try to detect the small amount of residual ^{244}Pu which might remain from the last nucleosynthesis in our solar system. She says a pre-Cambrian bastnasite (a rare earth fluorocarbonate mineral) which was being processed by the Molybdenum Corporation of America was chosen because CeO_2 is enriched in this ore by about 5×10^5 over its average terrestrial abundance. Plutonium (IV) would be expected to follow the Ce(IV) chemistry. The di-2-ethylhexylorthophosphoric acid (HDEHP) extractant was obtained after their process for

reduction and removal of cerium (which would not reduce and remove plutonium). The subsequent plutonium separations were performed by Hoffman and coworker Francine Lawrence in a new laboratory at the Los Alamos Scientific Laboratory utilizing "clean" room techniques (Fig. 2.7). The separations involved processing large quantities of the HDEHP extractant furnished by the Molybdenum Corporation, as well as running numerous blanks on all of the reagents and tracers used. Because of the extremely long half-life of ^{244}Pu (~80 million years), sensitive mass-spectrometric analyses were used for its measurement rather than trying to measure its very low specific activity α -decay. After the laborious chemical separations and processing by Hoffman and Lawrence, the final samples were sent to Jack Mewherter and Frank Rourke at the General Electric Co., Knolls Atomic Power Laboratory in Schenectady, New York, because they had one of the most sensitive mass spectrometers in the world

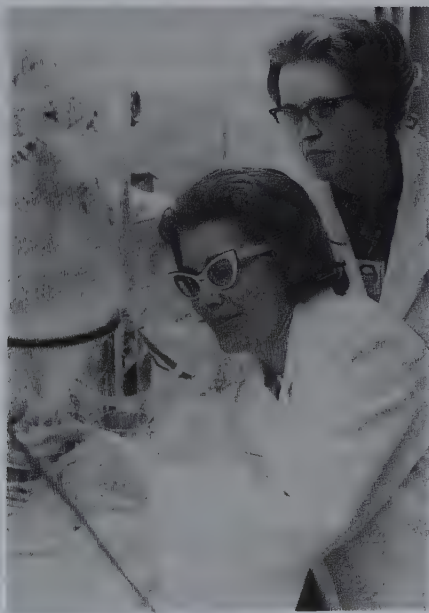


Fig. 2.7. Darleane Hoffman and Francine Lawrence, processing HDEHP to remove plutonium fraction, 1971, Los Alamos Scientific Laboratory.

for measurements of plutonium isotopes. Furthermore, they had much experience in measuring very low levels of plutonium in a wide variety of samples. Jack Mewherter performed the very careful final cleanup of the samples for the subsequent mass-spectrometric analyses performed by Frank Rourke.

In Darleane's words, "I recall being at the Santa Fe Opera on the Friday night in the summer of 1971 just after the final samples had been finished and as I looked out at the bright stars in the clear New Mexico sky behind the stage, I somehow had the feeling that this time we would find remnants of the elusive ^{244}Pu remaining from the last *r*-process nucleosynthesis of heavy elements in our galaxy some five billion years ago. And, indeed, after a two-year search, two aliquots of this final processing did show unmistakable evidence for ^{244}Pu . Ultimately, after careful analysis of all the data, including isotope ratios which ruled out the possibility of fallout or other contaminants as a source, we reported that 2×10^7 atoms (8×10^{-15} g) of ^{244}Pu had been detected in the plutonium fraction from extractant representing about 85 kg of ore containing 10% bastnasite. This corresponded to a concentration of about 10^{-18} g of ^{244}Pu per gram of pure bastnasite and implies a not unreasonable enrichment relative to cerium of 6 to 300, depending on what is taken for the ratio of ^{244}Pu to ^{238}U some 4.5 billion years ago. Thus it seems most probable that it is a remnant from *r*-process nucleosynthesis occurring at the time of formation of our solar system, although the possibility of accretion from a steady state cosmic ray source cannot be excluded." These results were published^{2,11} in the November 19, 1971, issue of *Nature*, but Glenn Seaborg, Chairman of the AEC until August 1971, actually made the first announcement of the discovery on September 9, 1971, at the 4th Geneva Conference on the Peaceful Uses of Atomic Energy. He had been following the progress of the work since he and Darleane had discussed it at a meeting at the Argonne National Laboratory in July 1971 commemorating Argonne's 25th anniversary. Darleane says she still has a letter in her files, thanking him for making this announcement and sending him a preprint of the *Nature* article. On her copy of the letter to Dr. Seaborg, the original

"Sincerely yours" at the end was replaced on Darleane's copy by her secretary to say "Love and kisses," presumably to tease her because she had been in telephone contact with Glenn concerning progress of the work and even visited him in his Washington office.

Darleane regrets that more follow-up research to determine whether ^{244}Pu has been concentrated in other ores, in deep sea sediments or manganese nodules, has not been carried out which might shed more light on whether it comes from a primordial or cosmic ray source. However, the research is particularly time-consuming and painstaking, and demands very special facilities, equipment, and expertise which typically cannot be devoted to such long term projects!

It is somewhat ironic that ^{244}Pu , the longest-lived known isotope of plutonium, was produced and discovered by "chance" by humans in their first thermonuclear explosion, "Mike," presumably by rapid capture of six neutrons in ^{238}U followed by β -decay. This process is similar to the r -process in nucleosynthesis and it is tempting to think that still heavier elements might also be produced in nature by similar processes, and many unsuccessful searches for SHE in nature have been made (see Chapter 14). However, because of the SF "disaster" which occurs in the heavy fermium isotopes, the route may have to be more devious than simple neutron capture.

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Chapter 3

The Plutonium People

The following account by Glenn T. Seaborg is largely devoted to the wartime characterization of the properties of plutonium and the development of the process for its production and use, with emphasis on some 200 of the large number of people involved. (Brief biographies of these participants are included in the book *The Plutonium Story*.^{3.1})

3.1. The Metallurgical (Plutonium) Project

In order to understand the plutonium production problem, let us summarize the reactions of the two isotopes of uranium with neutrons:



One of the questions which posed itself immediately was the following: Is it possible, by use of the mixture of the uranium isotopes as it occurs in natural uranium, consisting of about 0.7% of ${}^{235}\text{U}$ and 99.3% of ${}^{238}\text{U}$ by weight, to cause a chain reaction to occur on a very large scale? If so, the extra neutrons produced in the fission of ${}^{235}\text{U}$ would be absorbed by ${}^{238}\text{U}$ to form the desired isotope, ${}^{239}\text{Pu}$, in a large quantity.

The other question which of necessity came up for immediate discussion was: Would it be possible to devise, in a reasonable period of time, chemical means for separating this ${}^{239}\text{Pu}$ from the uranium and from the tremendous fission product radioactivities due to the many fission product elements that would be present with it?

These were the two staggering problems which formed the basis of the Plutonium Project. Their solutions were to a large extent unrelated, and the development program in connection with each of these problems was, of course, in the hands of research people in different fields, i.e. physics and chemistry.

It is beyond the scope of the present discussion to give more than a brief description of the development of the chain-reacting units called "piles" for the production of plutonium. The ideas for this originated with E. Fermi and coworkers at Columbia, and E.P. Wigner and associates at Princeton University. Fermi and L. Szilard were the first to recognize that the proper balance in the absorption of the secondary fission neutrons could be obtained in a heterogeneous lattice arrangement of solid pieces of uranium distributed in a more or less regular pattern throughout a graphite base. The graphite serves to slow down the fast neutrons produced in the fission reaction without absorbing them appreciably, and the two reactions listed above take place predominantly with the slowed neutrons. The original graphite-uranium pile, which became the basis for the production unit, was the result of the famous initial pioneering work of Fermi, H.L. Anderson, W.H. Zinn, B.T. Feld, G. Weil, and their associates performed at the University of Chicago, where such work had also been carried out by S.K. Allison, H.W. Newson, and coworkers. The first successful operation of the chain reaction, opening the atomic age, took place on December 2, 1942, under the West Stands at the Stagg Stadium football field at the University of Chicago. This reactor was soon moved out to a specially constructed building at Argonne Forest, then known as "Site A," where further important tests were carried out. It was not planned as a source of plutonium.

The early work of the chemistry group at the University of California formed the basis for the later development of the process used in the separation of plutonium from uranium and fission products in the large manufacturing plants. This process was based

upon the use of two oxidation states of plutonium, and a great deal of the earliest work was concerned with the study of these states on the tracer scale.

Early in 1942, following a high level decision made in Washington, D.C., on December 6, 1941, physicists who had been working on the development of the chain reaction and chemists who had been working on the development of the separation processes for plutonium, assembled at the Metallurgical Laboratory of the University of Chicago. In time, other scientists — chemists, physicists, engineers, biologists, and medical people — assembled there, to solve problems related to those described above. Although the Metallurgical Laboratory (the forerunner of the present Argonne National Laboratory) was the center of the activity, there were more than 70 research groups in various parts of the country that participated in the "Metallurgical Project" (a code name for the "Plutonium Project"). This entire Project was under the able direction of A.H. Compton. About 5,000 persons were engaged at the peak, of whom about 2,000 were at the Metallurgical Laboratory in Chicago. N. Hilberry was Assistant, or Associate, Director of the Project throughout most of its history. Allison also served as Associate Project Director during part of the early period. R.L. Doan, and later Allison followed by F. Daniels, served as Director of the Metallurgical Laboratory, while J.C. Stearns, R.S. Mulliken, and Z. Jeffries gave valuable administrative and advisory help.

Groups of physicists devoted to finding the best conditions for the production of the uranium chain reaction were headed by Fermi, Allison, and M.D. Whitaker. Wigner, G. Young, J.A. Wheeler, and others made important contributions to the design of the production reactors, which, on the basis of their early ideas, were soon visualized and finally built as water-cooled, aluminum-sheathed, uranium slugs embedded in a very pure graphite moderator. Szilard helped in all phases of the problem. Although it is beyond the scope of the present account to describe the work on the physics, engineering, and other aspects of the plutonium production problem, it should be mentioned that important contributions were made by L.B. Borst,

G. Breit, J.C. Chipman, E.C. Creutz, A.J. Dempster, A.B. Greninger, J.P. Howe, D.J. Hughes, M.C. Leverett, J.H. Manley, J. Marshall, L. Marshall, T.V. Moore, H.W. Newson, H.D. Smyth, A.H. Snell, E.S. Steinbach, E. Teller, H.C. Vernon, W.W. Watson, A. Wattenberg, G. Weil, A.M. Weinberg, and V.C. Wilson. H.N. McCoy helped in the organization of the Chemistry Division. R.S. Stone, with the help of S.T. Cantril, had supervision over the problem of radiation protection, and they were aided at the various sites by K.S. Cole, G. Failla, J.G. Hamilton, L.O. Jacobson, H.M. Parker, E.O. Wollan, and others. The important problem of the reduction of the uranium to very pure uranium metal was solved satisfactorily in an amazingly short time by F.H. Spedding and his associates, I.B. Johns, Jr., W.H. Keller, and H.A. Wilhelm, all at Iowa State College, and also by C.J. Rodden and his coworkers at the National Bureau of Standards, with the production of important smaller amounts by J.W. Marden and associates of the Westinghouse Company. J.R. Ruhoff and the Mallinckrodt Chemical Works are to be credited with the production of the pure uranium salts needed as a starting point for the production of the metal in quantity. The procurement of sufficient quantities of pure graphite, a very difficult problem, was solved through the efforts of Hilberry, Doan, and others; the quantities of high purity material needed for the first reactor at Chicago were furnished by the Speer Carbon Company and the National Carbon Company.

The Metallurgical Project was initially the responsibility of a committee headed by J.B. Conant and L.J. Briggs as a part of V. Bush's wartime Office of Scientific Research and Development. The Project became a part of the U.S. Army Manhattan Engineer District, known as the "Manhattan District," in September 1942. As is well known, the Manhattan District, which also took on similar responsibility with respect to the production and use of ^{235}U , was operated under the overall direction of General Leslie R. Groves, with the able assistance of Colonel K.D. Nichols, until after the end of the war, when the Atomic Energy Commission took over. Major A.V. Peterson had special responsibility with respect to the Metallurgical Laboratory.

The DuPont Company agreed late in 1942 to take on the responsibility for the building and operation of the plutonium production plant, which would be built, it was finally decided, at Hanford, Washington, to be preceded by a pilot plant at Clinton, Tennessee. From somewhat before this time until the successful completion of the job, hundreds of DuPont scientists and engineers became a part of the Project. Initial negotiations with DuPont involved C.M.A. Stine, E.K. Bolton, T.C. Gary, and R. Williams of that company. Much of the top direction of the DuPont effort was centered in Williams and in C.H. Greenewalt for the technical side of the job, in R.M. Evans for production, and in G.M. Read for design and construction. Leaders on the technical side of the effort included G.D. Graves, B.H. Mackey, J.N. Tilley, L. Squires, J.E. Cole, H. Worthington, W.E. Kirst, and J.B. Miles.

Thus it can be seen that the organization of the Metallurgical Project and the Metallurgical Laboratory and the associated efforts at other sites was a complex matter. Much of the discussion which follows will serve to illustrate the interrelationships between the various sites. Although the organization changed with time, the chart shown in Ref. 3.1, p. 237, dated January 23, 1943, is good for illustrative purposes, because it lists the key administrative people based at Chicago at a time when the Project was in full swing and before the departure of a number of these to other sites.

The responsibility for devising the chemical means of separating the ^{239}Pu from the uranium and fission products, as well as solving numerous other chemical problems related to obtaining a successful chain reactor for the production of the ^{239}Pu , rested with the Chemistry Division of the Metallurgical Laboratory. This division was successively under the direction of F.H. Spedding, S.K. Allison, J. Franck, T.R. Hogness, and F. Daniels. W.C. Johnson or P.W. Selwood served as associate director and L.B. Arnold, Jr., as assistant director during part of the time. The members of the division occupied temporary quarters in the Jones and Kent Chemical Laboratories of the University of Chicago during 1942. The group which Seaborg directed, with the assistance of I. Perlman, in the

research on the plutonium extraction process, occupied quarters which were extremely limited in size (one large and several small rooms at one end of the fourth floor of the Jones Laboratory). In December 1942, the group working on the plutonium extraction process and related problems, and the groups working on the fission products (under the leadership of C.D. Coryell), effects of radiation on critical materials and processes (under M.B. Burton), and analytical and other problems (under G.E. Boyd) moved to new quarters, the "New Chemistry" building on the edge of the campus. However, the space situation was soon critical again and an addition to this building, the "Chemistry Annex" or "New New Chemistry" building, was added late in 1943. (These buildings on Ingleside Avenue were torn down a few years after the end of the war.)

It should be added that the problem of assembling the large number of chemists and chemical engineers that were eventually needed at the Metallurgical Laboratory, many of whom went to the Clinton Laboratories and/or Hanford, was a very challenging one. A facility with the methods of radiochemistry was needed in most instances, and the available supply of trained radiochemists was essentially nil. Thus it was necessary to train scientists by the hundreds in the fundamentals and the techniques of this discipline. In order to build up Seaborg's staff, for example, he called on most of the competent scientists in his acquaintanceship to join him, and these in turn called on their friends. Thus his group eventually included a large fraction of his fellow chemistry students who had attended the University of California at Los Angeles as his classmates, in addition to many of his colleagues from the University of California at Berkeley. Seaborg gave a lecture course in nuclear chemistry during the summer of 1942, and the notes covering this course, taken by C.D. Coryell, were reprinted by the thousands.

During the first months at the Metallurgical Laboratory, intensive effort was directed toward devising the chemical separation process to be used in the production plants which were then being planned. Among the possible procedures that were investigated were precipitation, solvent extraction, volatility, adsorption-elution, and

pyrometallurgical and pyrochemical processes; in fact, all of the types of processes used later were included in this early scouting program of the year 1942. The solvent extraction approach was investigated by D.E. Koshland, Jr., the volatility approach by H.S. Brown and O.F. Hill and later E.G. Bohlmann, pyrochemical and pyrometallurgical procedures by T.T. Magel and M. Cefola, and the adsorption-elution method by J.E. Willard and E.H. Turk as well as Boyd, A.W. Adamson, E.E. Motta, E.R. Russell, J. Schubert, and their coworkers. However, it was decided before the end of 1942 to use a precipitation process, because this seemed to offer the greatest certainty of at least limited success in the short time interval involved, even though it did not seem to offer the greatest ultimate efficiency and would not lead to the recovery of the uranium for reuse.

The problem of separating the new element plutonium from uranium and fission products might not at first seem difficult, for it was, indeed, primarily a chemical problem. However, it differed in a great many ways from ordinary chemical problems, and these differences made the solution of the problem as a whole much more difficult, even though some of the differences actually helped the solution. From the beginning, the limited time seemed the most nearly insurmountable difficulty. It was by no means possible to complete the design and testing of the process before it had to be placed in operation. Even a simple chemical process usually requires a much longer time to place in large scale operation than did the plutonium separation process, even though the latter cannot be regarded as either simple or short.

The problem which had to be met during the fall of 1942 was that of developing a separation process which would meet the demanding requirements. The process had to accomplish a separation of plutonium in high yield and purity from many tons of uranium in which the plutonium was present at a maximum concentration of about 250 parts per million. Because of this low concentration, compounds of plutonium could not be precipitated, and any precipitation-separation process had to be based upon coprecipitation phenomena, i.e., the use of so-called "carriers" for plutonium. At the

same time, the radioactive fission products produced along with plutonium in the uranium (as a result of the fission of ^{235}U) had to be separated so that less than one part in 10^7 parts originally present with the plutonium would exist with the final product from the process. This requirement was necessary in order to make it safe to handle the plutonium, for without a separation of the fission products, the plutonium from each ton of uranium would have more than 10^5 curies of energetic gamma radiation associated with it. The process of separating fission products was called "decontamination." Thus a unique feature of the process was the necessity of separating a wide variety of elements completely from the final product and of accomplishing these separations by remote control behind large amounts of shielding in order to protect operating personnel from the hazards of the radiation. The finally decontaminated plutonium compound that was to be provided to those responsible for its ultimate use had to be one that would facilitate those final steps; it had to be a compound or solution of small bulk that could be shipped without difficulty, and it had to be of a composition that could be easily subjected to further purification. The separation process had to meet the further requirement that a "critical mass" of plutonium, which would lead to a disruptive nuclear chain reaction, should not accumulate at any step of the procedure.

If large amounts of plutonium had been available in the fall of 1942 and if its chemistry had been as well known as the chemistry of the more familiar elements, the task of developing the chemical process would still have been a formidable undertaking. Essentially all that was known about plutonium at this time was based upon secondary evidence from tracer experiments involving the infinitesimal amounts of the element which had been produced entirely by cyclotron bombardments. All of the plutonium (in the form of the isotopes ^{238}Pu and ^{239}Pu) used in the experiments up until the time of the operation of the Clinton reactor, with the exception of an experimental test of the separation process on the semiworks scale using some uranium oxide from the reactor at Argonne Forest, was cyclotron-produced, using the cyclotrons at the University of

California and Washington University. Tracer chemistry itself was a relatively new science; many of its phenomena were not clearly understood; and deductions based on it were often subject to doubt, particularly when applied to a new element. Added to the difficulty of devising the chemical process was the fact that only a few of the fission products had been identified, and many of these proved to be among the least-known of the chemical elements.

Operation of a chemical process by remote control behind massive shielding in an area which would become so highly radioactive after use that operating and maintenance personnel would not be able to enter the area for even brief periods of time to adjust and service equipment, made it imperative that the process be thoroughly tested in advance in the type of equipment to be used in the plant, in order to minimize the possibility of errors in the design of the process or the equipment. Furthermore, operational errors had to be kept to a minimum, and careful chemical control of the operations had to be maintained. In all of these considerations it was obviously desirable that the process to be operated should be as short and as simple as possible. Similarly, the process from the standpoint of plant design should consist of a number of similar steps requiring the same sort of equipment rather than consisting of steps which were so fundamentally different as to require many different types of equipment. At the same time, it seemed advisable to design the process and the equipment in such a way as to facilitate changes in case of failure. All of these requirements were met; in fact, the process was operated more successfully than even the most optimistic dared to hope and from the beginning gave high yields and decontamination factors.

Although it was felt that such a separation process would depend on the use of the two oxidation states of plutonium, which had been discovered during the early work at the University of California, the actual details, such as the best carrier compounds and best oxidizing and reducing agents, had not yet been discovered. S.G. Thompson is largely responsible for the conception and early development of the process which was finally chosen. The key to the process is the

quantitative carrying of plutonium (IV) from acid solution by bismuth phosphate, an unexpected phenomenon which was discovered in December 1942, and the expected noncarrying of plutonium (VI) by the same carrier material. This method, known as the bismuth phosphate process, operates as follows: neutron-irradiated uranium is dissolved in nitric acid, and, after the addition of sulfuric acid to prevent the precipitation of uranium, plutonium (IV) is coprecipitated with bismuth phosphate. The precipitate is dissolved in nitric acid, the plutonium (IV) is oxidized to plutonium (VI), and a by-product precipitate of bismuth (III) phosphate is formed and removed, the plutonium (VI) remaining in solution. After the reduction of plutonium (VI) to plutonium (IV), the latter is again coprecipitated with bismuth phosphate, and the whole "decontamination cycle" is repeated. At this point the carrier is changed to lanthanum (III) fluoride, and a similar "oxidation-reduction cycle" is performed, using this carrier, thereby achieving further decontamination and concentration. The plutonium at this point is sufficiently concentrated that final purification can be accomplished without the use of carrier compounds and plutonium peroxide is precipitated from acid solution.

3.2. Evolution of the Bismuth Phosphate Process

Because of the large number of workers who contributed to the development of the bismuth phosphate separation process over a relatively short period of time, it is difficult, if not impossible, to place in proper perspective all of the significant contributions. Similarly, efforts to give due credit to all who contributed to this work must involve many errors of omission. Since the conditions surrounding the developmental work were so abnormal, no one person could maintain intimate contact with all of the experimental details or be fully aware of all of the important decisions leading to the final successful operation of the process. This, therefore, is an attempt to delineate the chronological development of its major features.

One of the most remarkable features of the process is the limited time in which it was developed. The element plutonium was discovered in February 1941, and the first compound was isolated in August 1942, as will be described. The unusual properties of bismuth phosphate as a carrier for plutonium were discovered in December 1942, and the bismuth phosphate separation process was placed in successful operation in the pilot plant at the Clinton Laboratories in Tennessee in December 1943. Thus in actually less than one year the process development work was almost completed before the total quantity of plutonium available from cyclotron bombardments had reached 2 mg. Additional developmental work preceded the beginning of operation of the vast processing plants at Hanford, Washington, in December 1944. Finally, it is significant that only four-and-one-half years elapsed between the discovery of plutonium and its first use as a source of nuclear energy for military purposes. Accomplishment of this objective during World War II required not only tremendous financial resources, engineering and chemical skill, careful planning and organization, high priorities for material, skilled manpower, and construction facilities, but also imagination and (perhaps as much as anything else) a considerable measure of good fortune.

As a result of early work at Berkeley, one separation process had evolved already, based on the oxidation and reduction of plutonium and its nonprecipitation and precipitation with a lanthanum fluoride carrier. This was called the "lanthanum fluoride process," and it later became an integral part of the bismuth phosphate process. Although the lanthanum fluoride process seemed to be sound chemically, there were many potential difficulties inherent in its operation in the plant. In October 1942, the process was tentatively, albeit reluctantly, accepted as the best available prospect for use in the production plants which were to be constructed later, the design of which was already under consideration.

The early development of the lanthanum fluoride process pointed out the necessity of identifying the major fission products so that the chemistry of these elements could be taken into account in

working out the separation processes. Therefore, a brief survey of these fission products — their yields, half-lives, and other radiation characteristics — was made during August and September 1942, by B. Goldschmidt, a visitor from France, and I. Perlman. The results of these investigations were extremely useful and, in fact, crucial to the development of the separation process on the required short time scale. More complete information on the fission products became available later (too late to be useful in the development of the separation process) as a result of the work of C.D. Coryell and his group. The results of all this work were published under the names of the individual investigators, after the war, in the Plutonium Project Record.^{3,2}

In October 1942, the chemists at Chicago who were working on the chemistry of plutonium and attempting to develop the chemical process numbered about 20, but the number began to increase at about that time. The overall direction of the Metallurgical Laboratory research on the separation process for plutonium was Seaborg's responsibility. The investigation of various phases of the task was directed by Perlman, S.G. Thompson, J.E. Willard, G.W. Watt, and F.W. Albaugh, with groups led by J.L. Dreher, R.C. Thompson, D.G. Pye, and J.R. Gilbreath. (Numerous other contributors at this Laboratory and at other sites will be mentioned in later parts of this discussion.)

Separate groups of chemists were responsible for the identification and characterization of the fission products, for analytical problems, and for the study of the effects of radiation on the chemical extraction process and on critical materials. The fission product research was the responsibility of Coryell, with the help of N. Sugarman, A. Turkevich, W. Robinson, E.P. Steinberg, L. Winsberg, S. Katcoff, N. Elliott, H.A. Levy, D.N. Hume, E.L. Brady, R.R. Edwards, L.E. Glendenin, H.L. Finston, D.W. Engelkemeir, J.A. Swartout, N.E. Ballou, M.S. Freedman, R.P. Schuman, T.B. Novey, C.R. Dillard, W.H. Burgus, T.H. Davies, J.A. Marinsky, R.P. Metcalf, J.A. Seiler, and coworkers. G.E. Boyd, in charge of the analytical chemistry program, had the help of H.A. Potratz, M.S. Fred, R.E.

Curtis, J. Schubert, and others, while M.B. Burton, leader of the research on the effects of radiation, was assisted by A.O. Allen, T.J. Neubert, W.M. Garrison, A. Novick, E. Shapiro, C.V. Cannon, and others. By the summer of 1943, the chemistry division including these separate groups consisted of several hundred people and the project was expanding rapidly, although the chemists actually working on the separation process which was finally adopted numbered less than 25, excluding DuPont personnel. Sugarman replaced Coryell as leader of the remaining fission product research group, when the latter moved a large part of the group with him to the Clinton Laboratories. Similarly, E.B. Ashcraft and D.S. McKinney took over the responsibility for the analytical chemistry program when Boyd took over the leadership of this work at the Clinton Laboratories.

On the basis of these tracer experiments alone it was not possible to conclude that bismuth phosphate would carry plutonium at the relatively high concentrations expected to prevail in the large scale separations plant being planned for operation at Hanford. However, the first ultramicroscale experiments performed very soon thereafter by B.B. Cunningham, using methods described below, showed that bismuth phosphate was capable of carrying plutonium at concentrations up to and above those anticipated for actual plant operations.

During this period of early development of the bismuth phosphate process, little attention was given to the new process by the chemical engineers, although they were kept informed as to the progress of its development, because the decision had been made to develop equipment for a precipitation process adaptable to any similar system. Meanwhile, they had constructed a semiworks which was simply a small scale pilot plant composed of the same general type of equipment as was planned for use in a large plant later. The purpose of the semiworks was to test a process, especially the mechanical steps, under as nearly as possible the same conditions as would be expected to prevail in a large scale plant. In this way many difficulties with the process could be detected in time to make corrections in either the equipment or the chemical steps before building the plant and starting operation. The design and develop-

ment of the separations process semiworks at Chicago was done largely by a small group of men on loan from DuPont. The original group of engineers, which arrived in August 1942, operated under the supervision of C.M. Cooper, and included R.S. Apple, L.C. Peery, P.S. Vincent, D.S. Webster, F.B. Vaughan, and also W.A. Rodger from the Parke-Davis Company (Fig. 3.1). W.Q. Smith came a few months later to aid in the direction of this effort, along with M.D. Peterson and J.A. Lane, all from the DuPont Company. All of these men worked closely with the chemists, often with them in the laboratories. Their contribution to the development of the process was considerable, particularly in relation to its adaptation to large scale equipment and the suggestion of methods of practical improvement.

It was not until early in 1943 that the role of the DuPont company in the nuclear energy program became formalized. The project work was assigned to the Explosives Department in a new division known as TNX, and this assignment led to an expenditure of about \$350,000,000 for the establishment of the largest plant ever constructed and operated by the DuPont company up to that time.

Responsibility for the fundamental research and development essential to the success of the work done by DuPont remained with the University of Chicago. DuPont was asked first to engineer,



Fig. 3.1. Chemical engineers Luther Peery, Don Webster, with Dan Koshland (left to right) at Jackson Park, November 1942.

design, and construct a small pilot plant which was to be located near Clinton, Tennessee, but operated by the University. In addition, a large number of DuPont technical men were loaned to the University, which needed the skilled assistance provided by industrial experience.

As a result, DuPont began to have a major influence, beginning in February and March 1943, upon the development of the separations process. They began to evaluate the various possible processes which were being developed at that time. A group of process development chemists from DuPont was organized under the direction of J.B. Sutton at the Metallurgical Laboratory in January and February 1943, and included J.H. Balthis, Jr., C.M. Olson, J.H. Peterson, E.R. Gilbert, B.F. Faris, and G.W. Stahl. These men worked in an engineering division, which also carried other responsibilities and which, under Cooper, had already been in operation for a number of months. DuPont technical men soon became attracted to the bismuth phosphate process and in March and April 1943 began to put a major part of their effort into its development. They accomplished a great deal in the improvement of the process and in confirmation of the previous work of the process chemists working with Seaborg's group, which up to this time had been so small that less than six chemists could be spared for this work. Also, until this time, the chemical engineering group, which had been giving the major part of its effort to testing the lanthanum fluoride process, had not been able to expand sufficiently to put forth much effort in testing the bismuth phosphate process.

By May 1943 it had become clear that further concentration of effort was necessary to prepare the process for operation in the pilot plant at Clinton. DuPont personnel were particularly anxious that a single process be chosen, and a date was set for a meeting at which the decision would be made. On June 1, 1943, this historic meeting, attended by representatives of the DuPont Company and the Metallurgical Laboratory, together with the chemists and engineers who had been most closely associated with the development of chemical processes, was held in Chicago. All available information relative to

the bismuth phosphate and lanthanum fluoride processes was summarized. The decision which had to be made was not an easy one. Even in the light of all of the available data there remained uncertainties, for example, with regard to corrosion problems associated with the lanthanum fluoride process and to possible failure of complete coprecipitation of plutonium with bismuth phosphate at high concentrations of plutonium. In fact, it was not even certain that precipitation processes would necessarily prove superior to methods based upon solvent extraction, adsorption, or other phenomena. The decision in favor of the bismuth phosphate process was largely influenced by Seaborg's prediction of at least a 50% yield and by C.H. Greenewalt's expressed preference for a process that would minimize the possibility of early equipment failures even at the expense of decreased yields.

During the spring and summer of 1943, many experiments were performed with the purpose of defining more exactly the conditions for operating the process. During this period the work of the following men was particularly noteworthy: R.A. James, D.R. Miller, E.H. Turk, V.R. Cooper, J.L. Dreher, N.R. Davidson, D.E. Koshland, Jr., W.J. Knox, G.R. Leader, A.H. Angerman, and D.M. Ritter. Many experiments were tried in practically every case before suitable conditions could be established for each particular step.

The laboratory and semiworks tests had now proceeded to a point where, in the summer of 1943, the necessity for rapid development of methods for concentration and isolation of plutonium had become apparent. Such methods had been investigated in connection with the lanthanum fluoride process, but practically no work had been done on this phase of the bismuth phosphate process. In the laboratory, dissolution of bismuth phosphate in hydrochloric acid and the coprecipitation of plutonium with rare-earth-fluoride carrier had been extensively used and was remarkably satisfactory because of the high solubility of bismuth phosphate in hydrochloric acid. But hydrochloric acid was very corrosive to the stainless steel equipment planned for use in the process, and there appeared to be no simple method of avoiding this difficulty. It became necessary to choose

some concentration method which could be used in the semiworks equipment. It was at this point that the two precipitation processes which had been developed in parallel and which were both under consideration for use in the large scale plants (i.e., the bismuth phosphate and lanthanum fluoride processes) were combined, following a suggestion of L.C. Peery. The fluoride steps were used for the concentration of plutonium to reduce the bulk of the carrier following the several steps in which the bismuth phosphate procedure was used, and the later choice of an isolation procedure was to involve the precipitation of plutonium as the peroxide, a procedure proposed by I. Perlman for the lanthanum fluoride process. The real value of the combination did not become evident until later, when it became clear that fission products not well separated by the phosphate steps were efficiently separated by the fluoride steps. Furthermore, the use of the fluoride procedure was a protection against failure of the phosphate procedure, since in an emergency the former could have performed the extraction and decontamination steps.

The final step in the overall extraction and decontamination process was to be the isolation process, the precipitation of the plutonium in the form of a compound without the use of a carrier, after the concentration step. It was necessary to make a decision on the process for the isolation procedure before any of the many methods under investigation at the Metallurgical Laboratory and at Clinton had been perfected, in order that the design of the Hanford equipment might begin on time. Seaborg recalls being requested by Squires to select a process, even though no single one had yet been developed; he made the suggestion that it should be the peroxide precipitation procedure. Thus a single path was chosen, and the chemists were faced with the necessity of making a process work by one means or another. The very successful research that led to the perfection of the plutonium peroxide isolation procedure was carried out under the very able direction of G.W. Watt and D.G. Pye.

3.3. The Clinton Plant

It was agreed as early as September 1942 to locate the plutonium pilot plant in Tennessee. The move to the Clinton Engineer Works, or the Clinton Laboratories, named for the neighboring village of Clinton, took place during the fall of 1943. The plant had been largely built by that time and a nearby village, given the name of Oak Ridge, was constructed to house the personnel associated with the operation. M.D. Whitaker became the head of the plant; R.L. Doan, the director of research; W.C. Johnson, and later J.R. Coe, Jr., the head of the Chemistry Division; A.H. Snell, a leader of the Physics Division; M.C. Leverett, the head of the Technical Division; and K.Z. Morgan and J.E. Wirth, the heads of health physics and biology. Metallurgical Project Director Compton moved to the site to watch over the operations. L.B. Borst, L.W. Nordheim, and E.O. Wollan directed important segments of the work.

The Clinton Laboratories, also known by the code name of "Site X," was the responsibility of the University of Chicago, with personnel from the DuPont Company playing the key role in the design, building, and operation of the air-cooled graphite reactor and the bismuth phosphate extraction plant which served as prototypes or pilot plants for the later Hanford operation. (After the war the Clinton Laboratories became the Oak Ridge National Laboratory, operated first by the Monsanto Chemical Company, soon to be followed by the Union Carbide Company.) S.W. Pratt managed these pilot production activities from the standpoint of the DuPont Company, with the help of W.C. Kay, and with L.K. Wyatt in charge of the reactor and F.B. Vaughan in charge of the chemical extraction plant.

During the period August through November 1943 most of the chemists and engineers associated with the work on chemical processes moved from the Metallurgical Laboratory to the Clinton Laboratories to prepare for the beginning of plant operations there. The chain-reacting pile that had been under construction since early in 1943 began to operate at low-power level on November 4, 1943.

Table 3.1. Organization Chart for Section C-II, Clinton Laboratories (early 1944).

I. Perlman — Section Chief	
S.G. English — Associate Section Chief	
Mrs. D.C. Silverman — Secretary	
<i>Group 1 — Process Research</i>	<i>Group 4 — Product Isolation</i>
D.R. Miller — Group Leader	V.R. Cooper — Group Leader
A.F. Clifford	B.W. Bailey
A.H. Gernapy	L.B. Farabee
J. Halperin	R.H. Firminhac
J.P. Hunt	L.H. Gevantman
G.L. Johnson	A.J. Green
W.J. Knox	F.L. Halbach
D.E. Koshland, Jr.	K.K. Kennedy
J.P. MacBride	M. Lindner
E.D. Rose	L. Spector
R.B. Scott	
C.W. Smith	<i>Group 5 — Basic Chemistry</i>
H.G. Van Buren	L.B. Werner — Group Leader
J.H. Zimmerman	J.R. Dam
	J.C. Kroner
	G.E. Moore
<i>Group 2 — Process Service</i>	<i>Group 6 — Product Isolation</i>
S.G. English — Group Leader	B.A. Fries — Group Leader
J.O. Blomeke	J. Barrick
E.G. Bohlmann	J.P. Manion
O.F. Hill	
O.E. Myers	
<i>Group 3 — Physical Measurements</i>	
C.J. Borkowski — Group Leader	
R.L. Butenhoff, J.H. Parsons	
R.A. Dandl	
J.K. East	
V.O. Flateau	
A.A. Jarrett	
R. Jones	

Its performance was excellent. Construction of the separations plant was nearing completion. The process semiworks had been transferred from Chicago and began to operate in September 1943 in a division under the direction of O.H. Greager. The group of process development chemists under J.B. Sutton transferred to the Clinton Laboratories in November. A large number of chemists transferred from the Metallurgical Laboratory and continued in process work under the direction of I. Perlman, with groups under the leadership of S.G. English, D.R. Miller, D.E. Koshland, Jr., V.R. Cooper, and B.A. Fries. Table 3.1 shows a summary of the organization and various groups that worked under Perlman at this time. R.W. Stoughton later headed a group working on ^{233}U . Other groups with C.D. Coryell (on fission product research, hot-laboratory operations, and process thermodynamics) and G.E. Boyd (on analytical chemistry and continued research on potential adsorption processes as possible alternative methods for the separation and decontamination of plutonium) were similarly transferred to Clinton, and they all worked in the chemistry division under W.C. Johnson. H.S. Brown, after initially directing research on volatility processes, served as assistant director of the Chemistry Division.

The first uranium from the pile entered the separations plant on December 20, 1943. By the end of January 1944, metal from the pile was being processed in the plant at the rate of one-third ton per day; by February 1, 1944, 190 mg of plutonium had been isolated; and by March 1, several grams had been produced. This and the following plutonium was of special importance to the Los Alamos Laboratory. The yield from the plant at the very start was about 50%, and by June 1944 it was between 80 and 90%.

One very important phase of the laboratory work which must be mentioned was the isolation of the plutonium produced in the pilot plant. Even though considerable concentration had been accomplished in the concentration steps of the plant process, and the amounts of plutonium now becoming available were very large compared to those available before, they were much too small to be handled by the plant. Since isolation, then, was essentially a laboratory-scale

operation, a group supervised by V.R. Cooper was organized and given responsibility for isolation of the plutonium from the separations process pilot plant. L.B. Werner made the final isolation and hence was the first man in the world to handle subgram and then gram amounts of pure compounds of plutonium. He was aided in this difficult task by B.A. Fries.

Up to this time relatively little attention had been given to the study of isolation methods suitable for use at the Clinton Works. In addition to lanthanum fluoride and plutonium, the material delivered to Cooper's group contained extraneous insoluble solids, oil and grease, interfering ions such as zirconium (IV), and other impurities concentrated in the fluoride cycles designed to provide for the concentration of plutonium. It is indeed remarkable that approximately 99% of the plutonium received by the isolation group was recovered in substantially pure form.

After the operation of the Clinton extraction plant, and with the use of some of the plutonium thus made available, a complete mechanical test of the Hanford process on the semiworks scale was carried out. This work was done by a number of the chemical engineers working with O.H. Greager and gave information of great value to the ultimate success of the process at Hanford. This work tested the bismuth phosphate process at Hanford concentrations of fission products as well as at Hanford concentrations of plutonium (requiring some 10 g of plutonium).

The successful first and crucial test of the process at the Hanford plutonium concentration had been made somewhat earlier at the Metallurgical Laboratory on the ultramicrochemical scale, using cyclotron-produced plutonium, as described below. A further successful test at this concentration of plutonium was made at the Metallurgical Laboratory on the liter scale of investigation, using Clinton-produced plutonium. Such tests of the carrying ability of bismuth phosphate for plutonium (IV), established as the oxidation state which was coprecipitated so efficiently in the tracer experiments, were very necessary in view of the lack of understanding of the mechanism by which the plutonium was incorporated into the

bismuth phosphate crystal lattice. It could not be isomorphous incorporation in the lattice because of the difference in oxidation states of the plutonium and bismuth and the different crystal structures of their phosphates. There were many who were convinced that the process could never work at plant concentrations, some backing their convictions with bets, and a number of those holding these opinions were not convinced until the day the process was successful in the Hanford production plant. The test at the earliest possible time, leading to the ultramicrochemical program of investigation, was vital in order that another process might be chosen in the event that the test should indicate failure.

3.4. Ultramicrochemistry

As mentioned above, although the outline of a chemical separation process could be obtained by tracer-scale investigations, the process could not be defined with certainty until the study of it was made possible at the actual concentrations of plutonium that would exist in the large-scale separation plants. Such a test was particularly necessary in view of the poor understanding of the mechanism by which plutonium (IV) is carried by bismuth phosphate and the skepticism of many that the carrying would be observed at the concentrations of plutonium that would exist in the Hanford plant. This test had to be carried out as soon as possible after the discovery of the bismuth phosphate process, and it was performed at the Metallurgical Laboratory in early 1943, following the isolation of Plutonium the previous fall.

The question, in the summer of 1942, was as follows: How could any separations process be tested at the concentrations of plutonium that would exist several years later in the production plants when, at this time, there was not even a microgram of plutonium available? This problem was solved through an unprecedented series of experiments encompassing two major objectives. First, it was decided to attempt the production of an actually weighable amount of plutonium by bombarding large amounts of uranium

with the neutrons from cyclotrons. It must be remembered that never before had weighable amounts of transmutation products been produced with any particle acceleration machine. Even extending this possibility to the limit, it was not anticipated that more than a few micrograms of plutonium could be produced. The second aspect of the solution of this problem involved the novel idea of attempting to work with only microgram amounts of plutonium but, at the same time, at ordinary concentrations. It was decided to undertake a program of investigation involving volumes of solutions and weighings on a scale of operations much below that of ordinary microchemistry.

By using extremely small volumes it was possible to arrange matters so that even microgram quantities could give relatively high concentrations, and by developing balances of the required sensitivity, micrograms were sufficient for quantitative gravimetric measurements. The field which embraces the chemical study of material on this minute scale of operation has been given the name "ultramicrochemistry" by P.L. Kirk. The pioneer investigations of Kirk and A.A. Benedetti-Pichler in the fields of quantitative and qualitative analysis on the microgram scale were of the greatest value in providing a background of manipulative techniques for the problem at hand.

One type of balance used in the weighing experiments is the Salvioni balance shown in Fig. 3.2, consisting of a very thin quartz fiber, fastened at one end to a solid base; at the other end is attached, at right angles, a small holder for the sample to be weighed. The balance is named for the Italian investigator E. Salvioni, who in 1901 used fine glass fibers to weigh small samples of odorous substances to 0.1 μg in order to determine quantitative limits of olfactory detection. The determination of the weight of the sample depends upon measuring the amount of bending of the quartz fiber arm, and the balance is usually calibrated by weighing solid residues of known weight obtained as the result of the evaporation of aliquot samples of solutions of known compositions.

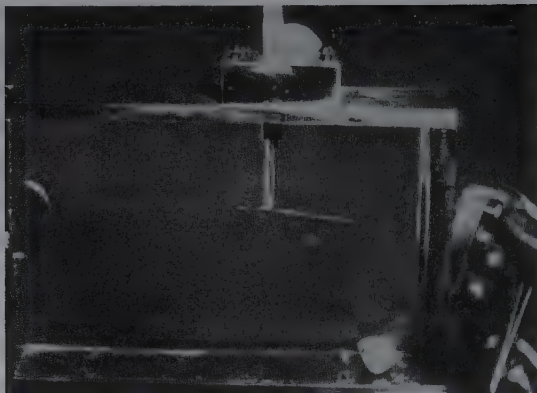


Fig. 3.2. Salvioni balance, with a sensitivity of $0.02\ \mu\text{g}$, a weighing range of $20\ \mu\text{g}$, and a capacity of $0.5\ \text{mg}$. It consists essentially of: a fine ($\sim 0.06\text{-mm}$ diameter) quartz fiber, $12\ \text{cm}$ in length, one end of which is rigidly mounted; an adjustable stop used in loading; an aluminum cradle for a weighing pan (weight $\sim 200\ \mu\text{g}$); and a platinum weighing pan (weight $\sim 150\ \mu\text{g}$). The position of the fiber is obtained from a reading microscope accurate to about $0.002\ \text{mm}$.

Another type of ultramicrobalance used in a number of these investigations had an extremely high sensitivity and was designed and built by Kirk, R.D. Craig, and J.E. Gullberg of the University of California. The design of the Kirk–Craig–Gullberg balance was influenced to a considerable extent by the early work on ultrasensitive microbalances by the Australian scientists B.D. Steele and K. Grant, and the work of the Swedish physicist H. Pettersson. This balance could weigh amounts as small as one microgram or less with an accuracy of $0.02\ \mu\text{g}$. In working with this balance and the small amounts of material which were weighed, it was said that “invisible material was being weighed with an invisible balance”!

3.5. Isolation of Plutonium

The isolation of plutonium at the Metallurgical Laboratory, after initial contributions by M. Cefola, was carried out chiefly by B.B. Cunningham and L.B. Werner, who had previously been occupied in

the biochemical field. The first pure chemical compound of plutonium, free from carrier material and all other foreign matter, was prepared on August 20, 1942, after starting with a plutonium concentrate in about 10 mg of rare earths prepared by A.C. Wahl and coworkers at Berkeley. This historic day marks man's first sight of the element plutonium and, in fact, the first sight of a synthetically produced isotope of any element. The excitement throughout the laboratory, which was situated in a guarded area on the fourth floor of the Jones Chemical Laboratory on the University of Chicago campus, was great indeed, as person after person came to view the historic sight under the microscope. Figure 3.3 shows a photograph of the room where this work was done; it had previously served as a balance room and was situated just off the larger laboratory, room No. 404.

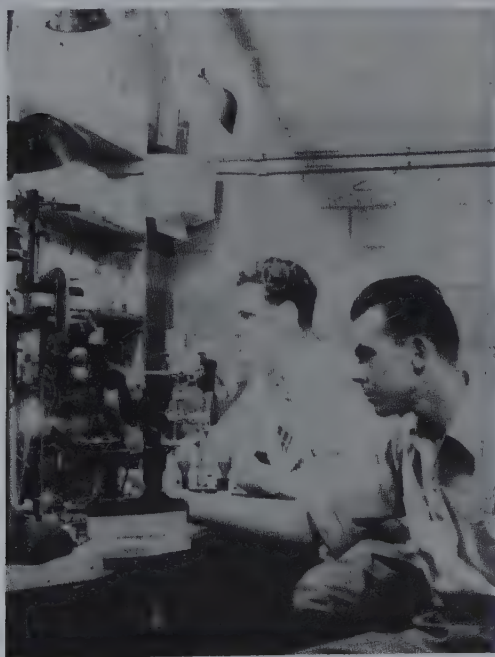


Fig. 3.3. B.B. Cunningham (*right*) and L.B. Werner, room 405, Jones Laboratory, University of Chicago, August 20, 1942.

It should be noted that at this time the element was often referred to as "element 94," or simply as "94." The name "plutonium" had been given it only a few months earlier and was not yet in general use on the Project. Also, it might be mentioned that the code name "49" was by this time being used to designate the isotope $^{239}\text{94}$, following the general (not very subtle) system which used the last digits of the atomic and mass numbers to represent an isotope.

The first weighing of a pure compound of plutonium occurred on September 10, 1942, when 2.77 μg of the oxide (PuO_2) were weighed by Cunningham and Werner. It was part of the plutonium produced in the large undertaking of the summer of 1942. A picture of this original oxide — carefully preserved and now on exhibition at the Lawrence Hall of Science at the University of California, Berkeley — is shown in Fig. 3.4. A second, 4.45- μg sample was used for a specific activity measurement.

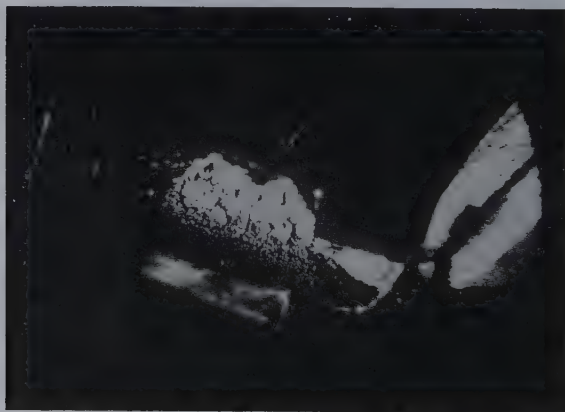


Fig. 3.4. The first plutonium compound (2.77 μg of oxide) to be weighed by man (September 10, 1942) is shown here on a platinum weighing boat. The picture is magnified approximately 35-fold. The plutonium oxide appears as a crusty deposit (indicated by the arrow) near the end of the platinum weighing boat, which is held with forceps that grip a small handle (upper part of photograph).

During this early period when visible amounts of plutonium were being precipitated, numerous visitors came to the laboratory to have a look at the fascinating new element. With the supply so limited and the experiments to be performed so many, it was not possible to keep much material immobilized to show them. Some of the hydroxide samples which were placed on view consisted of the much more available aluminum hydroxide colored with green ink; it should be understood, however, that great care was taken on all such occasions to remark, "*This represents* a sample of plutonium hydroxide," a statement which was often misunderstood to mean that it was actually a sample of plutonium hydroxide which was on display. One especially auspicious occasion found General Leslie R. Groves in the laboratory looking at the valuable new substance. The sample, in this case the real and precious plutonium hydroxide, was lined up under the microscope with the greatest of pride, and the disappointment was very great when he made the remark "I don't see anything."

During the summer of 1942, large amounts of uranium (hundreds of pounds) were bombarded for several months with the neutrons from the cyclotrons of the University of California, Berkeley, under the supervision of J.G. Hamilton; and Washington University, St. Louis, under the supervision of A.S. Langsdorf, Jr. The initial large-scale operations at the Metallurgical Laboratory for the isolation of plutonium were carried out by T.P. Kohman and A.H. Jaffey and coworkers. In these operations diethyl ether was used to separate the bulk of the uranium, as uranyl nitrate hexahydrate, from the plutonium (IV) and fission products; it is the success of this method that led to the above-mentioned consideration of solvent extraction as a possible chemical procedure to be used in the manufacture of plutonium. The lanthanum fluoride oxidation-reduction procedure was used for the separation of the plutonium from the fission products and for its concentration. These operations resulted in the production of several hundred micrograms of plutonium, a great

deal more than had been anticipated. This was used in the tracer-scale investigations of the chemical separation processes for plutonium and other tracer-scale investigations of plutonium, and also made it possible to carry out the program of ultramicrochemical investigation at Chicago (including the work just described above). In September 1942, B.B. Cunningham and L.B. Werner were able to prepare a number of compounds of pure plutonium and to determine with certainty, by means of chemical analysis, that the oxidation number of the most stable state of the element in solution is the IV state. M. Cefola, R.L. Patton, and C. Smith also made contributions to the program at the Metallurgical Laboratory, at this time as well as somewhat later.

The group remaining in Berkeley with W.M. Latimer also contributed to this ultramicrochemical program of investigation. During the summer of 1942, A.C. Wahl, independently, was also processing cyclotron-irradiated uranium in order to isolate pure plutonium. He succeeded in isolating 200 μg of chemically pure plutonium in 92% yield from 45 kg of uranium that had been irradiated for two months with neutrons from the Berkeley 60-Inch Cyclotron. He employed the lanthanum fluoride process and measured yields and decontamination factors at every step, collecting data which proved very valuable in the evaluation of this separations process, then the only practical method for isolation of plutonium. The chemistry was started in July, but progress was slow because of the care exercised in the evaluation of the separations process, and a pure compound of plutonium, namely plutonium (IV) hydroxide, was not isolated until September 29, 1942. However, it was with great elation that Wahl showed the 0.2-mg plutonium sample, easily visible to the naked eye, to E.O. Lawrence, whose cyclotron had produced the plutonium. This plutonium was used in an ultramicrochemical program of investigation at Berkeley, by Wahl, J.W. Gofman, J.W. Hamaker, G.E. Sheline, and W.H. McVey. Among other accomplishments, this group was able to establish in 1943 that the oxidation number of the highest state is VI.

From this time until the fall of 1943, cyclotron bombardments were the sole source of plutonium, and over this period of time about 2,000 μg , or 2 mg, of plutonium were prepared. This material was used to maximum advantage by the ultramicrochemists to prepare compounds of plutonium and to measure properties such as solubilities and oxidation potentials. In particular, it was possible — and this was of inestimable importance — to test the bismuth phosphate process which was under consideration for use at Hanford. The various parts of the complicated separation and isolation procedures were tested at the Hanford concentrations of plutonium in the careful and crucial experiments performed by B.B. Cunningham, L.B. Werner, D.R. Miller, I. Perlman, and others. Without the possibility of these tests early in 1943, it is fair to say that this process, which went into use at Hanford and turned out exceedingly well, would not have been chosen.

It should be emphasized that the scale-up between the ultramicrochemical experiments to the final Hanford plant amounts to a factor of about 10^9 , surely the greatest scale-up factor ever attempted. In spite of these difficulties the chemical separations process at Hanford was successful from the beginning, and its performance exceeded all expectations. High yields and decontamination factors (separation from fission activity) were achieved in the very beginning and continued to improve with time.

3.6. The Hanford Plant

Northwest from the village of Pasco in southeastern Washington, the Columbia River makes a 90° bend around a flat, arid expanse of land, which is bounded on its other two sides by high hills. Within this 600-square-mile area lie the several plants that made up the plutonium-manufacturing Hanford Engineer Works, known during wartime by the code name "Site W." The site was chosen with especial care for the needs of this unique enterprise. The mild winters and negligible rainfall allowed unimpeded construction the year around. The mighty Columbia River provided in necessary

amounts the low-temperature water to cool the chain-reacting piles. Flanking the area at convenient distances were the Grand Coulee and Bonneville power networks.

Not least among the considerations in the choice of site was the remoteness of the region from centers of population, a necessary forethought should unforeseen events have allowed the chain reactions to get out of control or the chemical plants to spew forth their contents, which were almost inconceivably high in radioactivity. It is a tribute to the painstaking care in planning that this massive undertaking offered the safest of industrial jobs to the operators.

The construction camp for the plant was built at the site of the village of Hanford, from which the plant drew its name. At one time over 50,000 people lived in trailers and barracks at Hanford, but with the completion of construction work it was fully evacuated. At a greater distance from the plant site a new village of permanent dwellings to house the plant operators was built at the location of the existing village of Richland.

There were two principal types of plants at the Hanford Engineer Works: those that formed the plutonium within the uranium by a nuclear chain reaction, and those that separated the plutonium from the uranium and the fission products. The former consisted of water-cooled piles, cooled by the circulation of treated Columbia River water. The huge separation units were strictly chemical plants, operated, like the piles, by remote control.

The Hanford site had been examined at the end of 1942 and was acquired by the government early in 1943. Construction of the piles began on June 7, 1943. By the spring of 1944 construction of the chemical plants was well under way and DuPont personnel were maintaining contact with developments at Wilmington, Chicago, Clinton, and Hanford. One of these men, L. Squires (who had participated in early process development work), kept himself closely informed of all developments at the various sites and expertly coordinated efforts in process improvement and equipment design. Blueprints of the equipment to be used at Hanford were constantly being submitted to representatives of the Metallurgical Laboratory

for examination and approval or suggestions for improvement. The canning problem (i.e., the problem of sealing the uranium in protective metal jackets) was a critical one. It was definitely not a simple matter to find a sheath that would protect uranium from water corrosion, keep fission products out of the water, transmit heat from the uranium to the water, and at the same time not absorb too many neutrons, but the problem was solved.

Prominent in the design of the various parts of the Hanford plant were R.P. Genereaux, F.W. Pardee, and S.L. Handforth, while F.H. MacKie, G.P. Church, and R.K. Mason were leaders in the construction phase. W.A. Dew was responsible for training the Hanford production personnel. W.O. Simon was plant manager during the startup and initial operation, and among his staff were such able people as D.O. Notman, B.H. Mackey, S.J. Bugbee, E.E. Swenson, W.C. Kay, L. Squires, F.A. Otto, and M.H. Smith. During the spring and summer of 1944 many technical personnel began to move to the Hanford site. These again included physicists, chemists, engineers, and others needed to ensure the success of the operation in a Technical Area. C.W.J. Wende, P.F. Gast, W.R. Kanne, and others played important roles in the field of physics. W.P. Overbeck made important contributions to the development of many of the electrical instruments that were needed. D.M. Smith headed an analytical division, and R.E. Curtis led a development group in this division. P.L. Kirk and coworkers developed methods for microchemical analysis of control samples, thus avoiding some of the hazards of working with large samples of the extremely radioactive process solutions. N. Hilberry, I. Perlman, and J.P. Howe represented A.H. Compton and the Metallurgical Laboratory, while H. Worthington, W.K. Woods, T.B. Drew, and J.A. Wheeler represented C.H. Greenewalt and the DuPont Wilmington office in connection with important liaison and advisory work concerned with critical parts of the program.

The chemical process investigations in the Technical Area were under the direction of J.E. Willard. Willard's organization was recruited from DuPont, Metallurgical Laboratory, and Clinton

Laboratories personnel and was composed of chemistry groups under the supervision of J.H. Peterson, S.G. Thompson, D.R. Miller, C.M. Olson, W. H Sullivan, and G.W. Watt. An organization chart for the groups working under Willard, corresponding to a period late in 1944, is shown in Table 3.2. Plant assistance groups under O.H. Greager and M.F. Acken included numerous chemical engineers who had had a long association with the process development work, e.g. R.S. Apple, R.H. Beaton, L.C. Peery, D.S. Webster, E.R. Gilbert, and J.B. Work. A semiworks group operated under the direction of I.D. Chambers and, later, W.W. Armstrong. In addition to the performance of assignments involving a continuation of process development work, the chief purpose of transferring these groups to Hanford was to provide expert advice and assistance in the event of serious chemical or operational difficulties at the Hanford plant. Similarly, personnel remaining at Clinton and Chicago provided a reserve of technical manpower available for diversion to work on Hanford problems in case of necessity.

Operation of the first pile began in September 1944, following some initial trouble caused by the absorption of neutrons in the fission product ^{135}Xe , and the first production run of Hanford material in a separations plant began on December 26, 1944. The final operating standards for the bismuth phosphate process, based on recommendations of the Clinton Laboratories, were reviewed at Hanford by a delegation from the Metallurgical Laboratory during December 12–15, 1944, and had just been finally approved on December 15, 1944. As had been true in the early operation of the pilot plant at Oak Ridge, a number of minor operational difficulties were encountered, and certain modifications in sampling methods and analytical procedures were required. The yields in the first plant runs, which took place in December 1944 and January 1945, ranged between 60 and 70%, reached 90% early in February 1945, and were up to 93% by early summer and above 95% soon thereafter. From the very beginning decontamination factors were better than anticipated and reached the overall value of 10^8 . The first delivery of plutonium to the Los Alamos Laboratory occurred on February 2, 1945.

Table 3.2. Organization Chart of Process Chemistry Section, Hanford Engineer Works (late 1944).

J.E. Willard — Area Supervisor

J.H. Peterson — Assistant Area Supervisor

Group 1 — Process Research

S.G. Thompson — Senior Supervisor

R.P. Black

J.L. Dreher

O.F. Hill

H.J. Kamack

W.J. Knox

H.T. Siefen

Group 4 — Product Isolation

G.W. Watt — Senior Supervisor

V.R. Cooper

B.A. Fries

R. Goeckerman

M. Lindner

D.G. Pye

F.R. Yett

Group 2 — Fission Products

W.H. Sullivan — Senior Supervisor

N.E. Ballou

T.P. Kohman

G. Leader

D.C. Lincoln

J.A. Swartout

Group 5 — Heavy Element Chemistry

D. R. Miller — Senior Supervisor

S.R. Gaarder

H.R. Hoekstra

W.C. Johnson

G.W. Sears

L.B. Werner

Group 3 — Process Development

C.M. Olson — Senior Supervisor

A.H. Angerman

B.F. Faris

W.L. Kay

B.H. Perkins

W.F. Schneller

G.W. Stahl

E.H. Turk

The chemical plants were massive structures ingeniously engineered to fit the grave problems inherent in handling the extremely high levels of radioactivity. It is self-evident that no one saw the plutonium as it entered the plant. It is also true that no one saw it until just before it finally emerged as a relatively pure compound.

In the meantime, the plutonium had passed through a maze of reaction vessels via thousands of feet of piping, with only instruments and an occasional sampling to chart its progress. Probably no process on a grand scale in chemical engineering history received such painful care in its development and engineering designs as that operating in the Hanford plutonium plant, certainly not in such a brief span of time.

Figure 3.5 is a picture of one of the chemical extraction plants at the Hanford Engineer Works in which the bismuth phosphate process was used. This view was taken right in the "canyon" area, where no one could be present when the process was operating. The actual operating cells containing the centrifuges and other apparatus are below the open area; the handles for lifting the covers to the concrete "cell blocks" can be seen. The remote control operating gallery is behind the thick concrete wall to the left. The crane which was used for the remote control lifting and replacing of equipment when the cell block covers were removed is shown above. The remote-control-operated cask car for the transport of material entered the canyon at the far right.

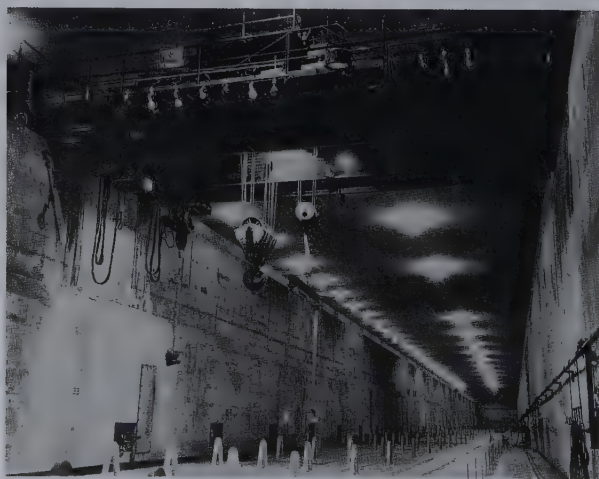


Fig. 3.5. View of Hanford Chemical Extraction Plant, in which the bismuth phosphate process was used.

3.7. The Los Alamos Laboratory

The Los Alamos Laboratory, known by the code name "Site Y," was operated (and still is operated) by the University of California. As is well known, the wartime director was J.R. Oppenheimer. Consideration of methods for constructing the weapon began as early as the summer of 1942 in Berkeley. The Los Alamos site for the Laboratory was chosen in November 1942, and by March 1943 Oppenheimer and some of the members of his research team had moved there. The Laboratory grew to the point where it included hundreds of scientists, recruited from all the other sites of the Manhattan District as well as from many other places. By 1945 this Laboratory had the highest priority with respect to personnel, as well as in other respects, and an extraordinarily capable staff had been assembled. At this time the organization included the following divisions: Experimental Nuclear Physics, under R.R. Wilson; Theoretical Physics, under H.A. Bethe; Chemistry and Metallurgy, under J.W. Kennedy, with C.S. Smith having special responsibility for metallurgy; Explosives, under G.B. Kistiakowsky; Ordnance, under Captain W.S. Parsons (U.S.N.); Bomb Physics, under R.F. Bacher; and Advanced Development, under E. Fermi. Fermi and S.K. Allison, who assisted Oppenheimer in the coordination of the research effort, had moved to Los Alamos from the Metallurgical laboratory in 1944 after the problems connected with the production reactors had been solved.

It was necessary to measure the fission and neutron capture cross sections of ^{239}Pu and the uranium isotopes, the elastic and inelastic neutron cross sections for numerous nuclei, the number, energy, and time of emission of the neutrons emitted in fission, spontaneous fission (SF) rates, and numerous other nuclear constants. It is not possible in the scope of this discussion to do more than mention some of the individual contributors. J.H. Manley was concerned with the measurement of critical neutron cross sections. J.H. Williams and H.H. Staub measured neutron fission cross sections and the energy spectrum of the neutrons emitted in fission. E. Segrè and D.E. Nagle

measured the ratio of neutron capture to fission in plutonium. R.R. Wilson made important measurements on the time of fission and other details of this process. R.W. Dodson, Segrè, and A.C. Wahl were concerned with the products formed by the irradiation of ^{239}Pu with neutrons. Of especial importance was the discovery by O. Chamberlain, G.W. Farwell, and Segrè (with later contributions by C.E. Wiegand) that ^{240}Pu , present in Hanford plutonium as the result of the absorption of neutrons in ^{239}Pu , undergoes SF at a relatively high rate.

Crucial questions, the answers to which the successful use of plutonium in a weapon depended upon, were whether ^{239}Pu emitted neutrons when it underwent fission, and the number of such neutrons emitted. These questions had to be answered early, before any ^{239}Pu was available from the reactors. Most of the supply of our chemistry group at the Metallurgical Laboratory was loaned in the summer of 1943 to Los Alamos for the purpose, and their initial measurement was made with this sample containing a few hundred micrograms. In returning the sample to the Metallurgical Laboratory, advantage was taken of my presence in Santa Fe on a short vacation trip in order to save the time which would have been lost in a cumbersome official transfer of the material. Wilson met Mrs. Seaborg and me at breakfast in a Santa Fe restaurant, having escorted the sample from Los Alamos with the implied protection of a high-powered rifle; I then transported it by train in my suitcase, without firearms, back to the Chicago Laboratory.

The Chemistry and Metallurgy Division was responsible for the final purification of the ^{239}Pu (and ^{235}U), for fabrication of the weapon, including the core and tamper, and for various other matters. People working under the direction of J.W. Kennedy, such as A.C. Wahl, C.S. Garner, R.B. Duffield, D. Lipkin, S.I. Weissmann, M.L. Perlman, and others, were principally concerned with Plutonium chemistry as applied to plutonium purification; these requirements were very stringent, especially with respect to light-element impurities. I.B. Johns, Jr., H.A. Potratz, and F.K. Pittman were among the others

who made important contributions. The remarkable feat of preparing pure plutonium metal and determining its important properties, using milligram and gram amounts, was accomplished under the direction of C.S. Smith, with E.R. Jette, R.D. Baker, M. Kolodney, E.F. Hammel, Jr., and others making important contributions. C.H. Thomas, with the help of J.C. Warner and J.H. Lum, acted as a coordinator and advisor in the plutonium chemistry and metallurgy program.

The Theoretical Division included a number of very able men, such as H.A. Bethe, R.F. Christy, E. Teller, R.P. Feynman, R. Serber, and V.F. Weisskopf. Among the problems considered were the determination of the critical size of the fissionable material in the weapon, the initiation of the explosion by stray neutrons, the rate of neutron density increase and other changes during the explosion, the equation of state of matter at high temperatures and pressures, and the general problem of how to proceed from data obtained with small quantities of materials to estimates as to what would happen in the weapon. The work of the Ordnance, Explosives, and Bomb Physics Divisions was, of course, concerned with the practical details of designing and building a practical weapon.

Thus it can be seen that the solution to the problem of transforming the plutonium, as received in solution from the Hanford Plant, into a workable weapon depended upon the work of many people in many different fields. Some of them, of course, were more concerned with the final design and assembly of the weapon and made their most important contributions to this final and most crucial stage of the development. The actual building of the plutonium weapon required many brilliant ideas of a fundamental type and important ideas for the details of design. Among those who made contributions in connection with the successful fabrication of the plutonium weapon were L.W. Alvarez, R.F. Bacher, K.T. Bainbridge, R.F. Christy, C.L. Critchfield, R.P. Feynman, L.H. Johnston, G.B. Kistiakowsky, E.M. McMillan, S.H. Neddermeyer, and R.E. Peierls.

3.8. Some Other Early Contributors

After the initial major emphasis on the separations process work, the research of the chemistry group at the Metallurgical Laboratory took on a broader aspect. Until the successful operation of the chain reactor and chemical separation plants, first at Clinton and then at Hanford, the work continued on the tracer scale and on the ultramicrochemical scale of investigation. After plutonium became available in larger amounts from these plants, it was possible to extend the scope of all of these investigations.

The group of chemists at the Metallurgical Laboratory at the University of Chicago continued to make basic solution and dry chemistry studies, under the direction of B.B. Cunningham, and early plutonium purification studies, under the direction of E.F. Orlemann. Further work on the plutonium separation and decontamination problem was carried out under the direction of S.G. Thompson, in groups under the leadership of F.W. Albaugh, J.L. Dreher, and G.W. Watt. This effort came under the direction of Albaugh when S.G. Thompson went to Hanford, and involved groups under R.C. Thompson, J.R. Gilbreath, and S. Lawroski. Investigation of the potentialities of U-233, which had been carried out under the direction of R.W. Stoughton, was continued in a group under the leadership of L.I. Katzin. Seaborg was assisted at various times in the direction of this entire effort by J.E. Willard, W.M. Manning, and G.W. Watt. (After the war, Manning became the Director of the Chemistry Division and continued to serve in that capacity in the Argonne National Laboratory.) The personnel changed continually as the people went to the various sites, so that it is difficult to give a short description of the problems which were studied and those who were involved. Perhaps the best summary can be given by showing the organization charts for two periods in the time interval between the early work described above and the end of the war. Table 3.3 shows such an organization chart corresponding to early 1944, while Table 3.4 shows one for about a year later.

Table 3.3. Organization Chart of Section C-1, Metallurgical Laboratory (early 1944).

G.T. Seaborg — Section Chief
 E.E. Smith — Secretary to Section Chief
 K. Buehler — General Secretary to Section
 W.M. Manning — Associate Section Chief
 G.W. Watt — Associate Section Chief
 J.E. Willard — Associate Section Chief
 L.I. Katzin — Assistant to Section Chief
 I.N. Saxton — Secretary to Associate Section Chiefs

Subsection I — Separations Processes

S.G. Thompson — Assistant Section Chief
 D. Gottlieb — Secretary

Group 1 — Extraction-Decontamination

F.W. Albaugh — Group Leader
 M. Ader
 L.S. Bartell
 R.H. Bradt
 C. Egan
 R.W. Greenlee
 H.R. Hoekstra
 J.G. Malm
 L.O. Morgan
 S. Peterson
 R.G. Post
 J. Sedlet
 M. Summers
 R.C. Thompson

Group 3 — Process Development

J.L. Dreher — Group Leader
 W.J. Blaedel
 P. Boykin
 J.E. Freeman
 J.R. Gilbreath
 H.H. Hyman
 R.G. Larson
 D.C. Lincoln
 A. Parnell
 R.W. Rasmussen
 B.M. Winner

Group 2 — Concentration-Isolation

D.G. Pye — Group Leader
 W.C. Beard, Jr., M.T. Walling
 R.H. Goeckerman, F.R. Yett
 F.W. Haeckl
 H.H. Hopkins, Jr.
 J.J. Katz
 A.E. Kelley

(Continued)

Subsection II — Purification and Metal Production

E.F. Orlemann — Assistant Section Chief

E. Kelly — Secretary

Group 4 — Purification and Analysis

L.H. Jensen — Group Leader

B.B. Brody

J.S. Dixon

S. Lawroski

R.A. Reinhardt

A.P. Stein

*Group 5 — Volatility and General
Dry Chemistry*

N.R. Davidson — Group Leader

B.M. Abraham

F.T. Hagemann

E.K. Hyde

H. Pellock

I. Sheft

M.J. Wolf

Group 5A — Fluoride Chemistry

R.E. Heath — Group Leader

A.E. Florin

F. Meyer

S. Nyden

H.P. Zvolner

Group 6 — Metal Production

H.L. Baumbach — Group Leader

R.D. Frank

S.M. Fried

M. Gerstein

N.N. Hellman

Z.V. Jasaitis

S. Katz

H.P. Robinson

E. Watts

E.F. Westrum, Jr.

Group 6A — High Vacuum

O. C. Simpson — Group Leader

N.D. Erway

L. Gilpatrick

F. Johnson

T.E. Phipps

G.W. Sears

Subsection III — Basic Chemistry and Service

B.B. Cunningham — Assistant Section Chief

M. Bohlman — Secretary

Group 7 — Basic Chemistry

J.C. Hindman — Group Leader

D.P. Ames

H. Billington

J.J. Howland

R.A. James

Group 8 — Recovery

L.R. Dawson — Group Leader

H.H. Anderson

L.B. Asprey

N.S. Baumbach

J.W. Britain

(Continued)

K.A. Kraus
T.J. La Chapelle
C.K. McLane
L.B. Magnusson
P.R. O'Connor
C. Smith

J.G. Burr, Jr.
P.R. Fields
P. Fineman
L. Leventhal
D.C. Stewart
M.H. Studier
D.B. Wetlaufer

Group 9 — Instruments and Physical Measurements

A. Ghiorso — Group Leader

J.A. Crawford
L. Golden
D.L. Hufford
A.H. Jaffey
T.P. Kohman
A.C. Krueger
E. Lewis
B.F. Scott
P.D. Walsh
B.B. Weissbourd

Table 3.4. Organization Chart of Section C-1, Metallurgical Laboratory (February 1, 1945).

G.T. Seaborg — Section Chief	
E. Albaugh — Secretary to Section Chief	
W.M. Manning — Associate Section Chief	
J. Horwich — Secretary	
T.O. Jones — Assistant to Section Chief	
M. Williams — Secretary	
K. Florin — Supervisor of Service Group	
<i>Subsection I — Separations Processes</i>	
F.W. Albaugh — Assistant Section Chief	
D. Black — Secretary	
<i>Group 1 — Extraction-Decontamination</i>	<i>Group 4 — Solvent Extraction</i>
R.C. Thompson, Jr. — Group Leader	S. Lawroski — Group Leader
R.H. Bradt	C.J. Egan — Squad Leader
H.H. Hopkins, Jr.	I.J. Schaffner — Assistant Squad Leader
J. Krinsky	B. Murray — Typist
J.G. Malm	M. Ader
L.O. Morgan	G.J. Bernstein
S. Peterson	B.B. Brody
M. Summers	D.J. Dorcy
	E.A. Hausman
<i>Group 3 — Process Development</i>	H.H. Hyman
J.R. Gilbreath — Group Leader	A.E. Kelley
M. Bolden — SECRETARY	R.A. Reinhardt
W.C. Beard, Jr.	J.H. Schraidt
W.J. Blaedel	W. Simon
P. Boykin	B. Struminski
R.G. Post	
J. Sedlet	
M.T. Walling, Jr.	
B.M. Winner	

(Continued)

Subsection II — Basic Chemistry and Service

B.B. Cunningham — Assistant Section Chief

R. Rogers — Secretary

Group 5 — Basic Dry Chemistry

O.C. Simpson — Group Leader

N.R. Davidson — ASST. Group Leader

N.D. Erway

S.M. Fried

H. Peterson

T.E. Phipps

H.P. Robinson

R.L. Seifert

I. Sheft

H. Thomson

E.F. Westrum, Jr.

Group 8 — Instruments and Physical Measurements

A. Ghiorso — Group Leader

A.H. Jaffey — Assistant Group Leader

J.A. Crawford

J.M. Dorsey

D.L. Hufford

A.C. Krueger

E. Lewis

B.F. Scott

P.D. Walsh

B.B. Weissbourd

Group 6 — Basic Wet Chemistry

J.C. Hindman — Group Leader

D.P. Ames

H. Billington

J.S. Dixon

A.E. Florin

R.W. Greenlee

J.J. Howland

R.A. James

T.J. La Chapelle

L.B. Magnusson

C.K. McLane

P.R. O'Connor

Group 9 — 23 Work

L.I. Katzin — Assistant Section Chief

F.T. Hagemann

N.N. Hellman

E.K. Hyde

R.G. Larson

M. S. Potter

M.H. Studier

Q. Van Winkle

M.J. Wolf

Group 7 — Recovery

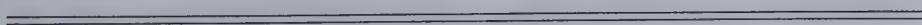
D.C. Stewart — Group Leader

H.H. Anderson, P. Fineman

L.B. Asprey, O.M. Giacchetti

J.W. Britain

P.R. Fields



In addition to the need for work with pure plutonium in connection with the separations process, it was necessary to determine a number of the physical and chemical properties of the dry salts of plutonium and of plutonium metal. Therefore, the study on the ultramicroscale had to encompass this field of investigation also. A number of compounds of plutonium were prepared by reactions involving the solid and gas phases — that is, by dry chemical reactions. This work was done in collaboration with W.H. Zachariasen and R.C.L. Mooney of the University of Chicago staff, who were able to use the X-ray diffraction technique to identify or help identify a number of the compounds that were synthesized and, in many cases, thus to establish their chemical structures. This preparative work and measurement of properties, as well as, later, the use of larger amounts of plutonium from the reactors, was under the direction of an extraordinarily able group of chemists whose accomplishment seems all the more remarkable in retrospect, considering the very limited amounts of material with which they had to work and their lack of previous experience in very-small-scale chemical manipulation.

The dry chemistry program was organized roughly along the following lines: (1) studies of the preparation and properties of the important classes of solid anhydrous plutonium compounds, such as oxides, halides, oxyhalides, sulfides, nitrides, etc.; (2) determination of the basic thermodynamics of such compounds; and (3) preparation and determination of the properties of plutonium metal. The very rapid progress achieved in carrying out the preparative portion of this program was due in large measure to two factors: the ingenuity and the experimental skill displayed by the chemists in devising methods for conducting dry-chemical reactions in tiny, thin-walled capillary tubes, and the astonishing ability of W.H. Zachariasen in securing and interpreting X-ray diffraction patterns on products so prepared. Thus it was possible in many cases to determine the results of an experiment within a few hours *without destroying the sample*. Had it been necessary to depend upon conventional microanalysis for identification of compounds, days and even weeks might have been required for each analysis.

The first preparation and identification of the important compound PuCl_3 was done by F.T. Hagemann in February 1944 and identified by Zachariasen by its X-ray diffraction pattern. The preparation of pure plutonium tribromide was achieved first by E.K. Hyde, and its identity was established by conventional chemical analysis.

Quite often, in working with very small amounts of material the products obtained were prepared inadvertently, due to the presence of impurities (especially traces of water and oxygen), and in such cases the X-ray diffraction method quickly revealed the true situation. It was in this way, for example, that plutonium oxyiodide was first obtained and identified.

In the early work on the preparation and identification of the plutonium fluorides A.E. Florin and R.E. Heath were the principal contributors, although the fluoride research later expanded to include many investigators, among whom were J. Karle, C.S. Smith, H.H. Anderson, N.R. Davidson, and S. Fried; L.B. Magnusson and T.J. LaChapelle did much of the work on neptunium fluorides. Work on the chloride, bromide, and iodide of plutonium was carried out by N.R. Davidson, J.J. Katz, E.K. Hyde, F.T. Hagemann, B.M. Abraham, B.B. Brody, I. Karle, M.J. Wolf, and I. Sheft. E.F. Westrum, Jr., made important investigations of the oxides PuO_2 , Pu_2O_3 , and PuO , while studies of the sulfides, oxysulfides, and nitrides were conducted by Abraham, Davidson, and Westrum.

In pursuing the second objective of research in the dry chemistry program — that of securing basic thermodynamic data on these substances — three general lines of approach were used: (1) direct thermochemical measurements, (2) observations of the temperature coefficient of equilibrium constants, and (3) rough estimates of lower or upper limits of stability by noting the presence or absence of reactions under chosen experimental conditions.

The success of the thermochemical program was due in very large measure to the efforts of Westrum and H.P. Robinson, who succeeded in constructing a heat-of-solution calorimeter of semimicro size and high accuracy. With this apparatus Westrum and Robinson determined standard heats of formation of plutonium aqueous ions,

and from these, of solid plutonium trifluoride, trichloride, tribromide, and oxychloride, using milligram samples.

Notable contributions to the plutonium thermodynamics program were made by Davidson, Sheft, and Fried, who investigated various halide and oxyhalide equilibria as a function of temperature. A special contribution to plutonium compound thermodynamics was that of O.C. Simpson, T.E. Phipps, G.W. Sears, and R.L. Seifert, who carried out a series of remarkably precise measurements of the vapor pressures of PuF_3 , PuCl_3 , and PuBr_3 as a function of temperature and also investigated the volatility of plutonium oxide at elevated temperatures.

In all of the work on plutonium thermodynamics done at Chicago, much benefit was derived from correspondence and occasional consultation with L. Brewer of the University of California, whose extensive knowledge of, and insight into, the field of thermodynamics served to clarify and, in some cases, to correct initial conclusions regarding the experimental data.

The third and perhaps most exciting phase of the early program on plutonium dry chemistry was that which had as its objective the preparation and determination of certain physical properties of plutonium metal, pertinent to its use in a nuclear weapon.

Following some unsuccessful attempts to make elemental plutonium by reacting the oxide with metallic potassium, H.L. Baumbach and P.L. Kirk in November 1943 made the first microgram samples of the metal by reducing 35 μg of plutonium tetrafluoride with metallic barium in a thoria crucible at 1400°C . The metal appeared in globules weighing about 3 μg each, and it had a silvery luster. It was proved to be the metal by the fact that it reacted rapidly with hydrogen to form plutonium hydride.

Using metal produced in such a system, Kirk and Baumbach, later joined by Fried, determined the density of their samples by dropping a weighed piece into a capillary tube of known diameter, partially filled with dibutylphthalate, an inert organic liquid of low vapor pressure. From the displacement of the liquid meniscus caused by adding the metal, the volume of the piece could be determined, and,

from this volume and the known weight of the sample, the density of the earliest samples was calculated to be 16 g/cm^3 . The density so determined (probably not that of a single pure phase of the metal) was later confirmed by W.H. Zachariasen by X-ray diffraction measurements. Using such metal Z.V. Jasaitis was able to determine the melting point, and R.S. Rosenfels developed methods for mounting the metal for hardness determinations and metallographic examination.

The ultramicroscale method of metal production described above was modified and improved by Fried and Westrum, who made major contributions to the program on metallic plutonium.

Later, at the time milligram amounts of plutonium became available for research, O.C. Simpson and his colleagues were able to get very precise measurements of the vapor pressure of the liquid metal over an extended range of temperature.

Worthy of special mention is the work of J.C. Hindman and coworkers on the solution chemistry of plutonium. They made many fundamental contributions to the elucidation of the ionic species present in aqueous solutions of different acids and to the determination of various complex ions of plutonium. K.A. Kraus also made many important contributions to these general problems. The overall contributions of B.B. Cunningham to this solution chemistry program, as well as to the above-mentioned dry chemistry program, are also noteworthy.

The problem of the purification of the plutonium to the stringent specifications required for its use as a weapon was, as mentioned above, the responsibility of E.F. Orlemann. Under his excellent direction many results, useful to the Los Alamos chemistry group in helping them meet their requirements, were obtained. Of especial importance was the work of Orlemann's group in developing various solvent extraction procedures for this purpose. Important contributions to the program were made by L.H. Jensen, J.I. Watters, B.B. Brody, R.A. Reinhardt, J.S. Dixon, and others. The development of analytical methods of importance to this program was done in a group under the leadership of H.A. Potratz. M.S. Fred and N.H.

Nachtrieb made valuable contributions to spectrographic analysis methods.

Also very important to the effort of the chemistry groups at the Metallurgical Laboratory was the work of the "Recovery Group," under the direction of L.R. Dawson and later under D.C. Stewart. This group had the important assignment of continuously recovering for reuse the precious plutonium as it found its way into peculiar places in the course of the experimentation program.

Problems of instrumentation and radioactivity measurements were in the capable hands of A. Ghiorso, A.H. Jaffey, T.P. Kohman, C.J. Borkowski, and H.P. Robinson, with the help of J.A. Crawford, D.L. Hufford, B.B. Weissbourd, B.F. Scott, and others.



Fig. 3.6. Met Lab Section C-I Reunion, July 14, 1979. *Standing, left to right:* Clark Egan, Stephen Lawroski, William Knox, Herman Robinson, Fred Albaugh, Edrey Albaugh, Harrison Brown, John Howe, J. Leonard Dreher, Leonard Katzin, Roy Greenlee (back), John Dorsey (front), Earl Hyde, Larned Asprey, Donald Wetlaufer, David Templeton, Walter Beard, Donald Pye, Harry Kamack, Sonia Katz, Lorraine Crawford, Ralph Firminhac, Quentin Van Winkle, Jack Tepe, Zene Jasaitis, Jonathan Dixon (back), Ted La Chapelle. *Middle row:* Al Ghiorso, Nancy Leverett, Marilyn Howe, Annis Moore, Marge Asprey, Helen Seaborg, Glenn Seaborg, Bernard Fries, Roy Beaton, Leon Leventhal, Roy Heath, Mary Ellen La Chapelle, Marguerite Dorsey. *Front row:* Alex Kelley, Miles Leverett, Al Florin, Kay Florin, Elwin Covey, Joe Katz, Elton Turk, Tom Morgan, Jerome Howland, John (Osty) Ostapowicz.

Figure 3.6 is a picture of a Met Lab Section C-I Reunion held on July 14, 1979, at the Seaborg home in Lafayette, California.

The group remaining at the University of California under the overall direction of W.M. Latimer, after the removal of J.W. Kennedy, A.C. Wahl, M.L. Perlman, C.S. Garner, R.J. Prestwood, D. Lipkin, S.I. Weissmann, R.B. Duffield, and others to Los Alamos in the spring of 1943, continued to make important contributions. R.E. Connick and a group consisting of W.H. McVey, G.E. Sheline, J.W. Hamaker, M. Kasha, M.W. Evans, E.L. King, and others made outstanding advances in the solution chemistry of plutonium, including the oxidation states and potentials and reaction kinetics. E.D. Eastman, L. Brewer, and a group consisting of L.A. Bromley, D.D. Cubicciotti, Jr., N.L. Lofgren, C.D. Thurmond, O.A. Cook, B.J. Fontana, M.J. Sienko, P.W. Gilles, and others contributed notably to the important problem of finding suitable container materials in which to carry out the reduction of plutonium fluoride to the metallic state, which was of crucial importance to the success of the Los Alamos task. G.K. Rollefson and H.W. Dodgen were the first to determine a number of the important lines in the plutonium emission spectrum. (R. Overstreet and L.O. Jacobson, with J.G. Hamilton, worked on the identification of a number of fission products.) J.W. Gofman and a group consisting of H.W. Crandall, G.C. Pimentel, W.H. Reas, and C.E. Crompton made contributions to the plutonium extraction and decontamination problem.

F.H. Spedding, together with A.F. Voigt and F.J. Wolter at Iowa State College, contributed to the chemistry of complex ion formation as well as to other phases of plutonium chemistry. (Here a number of important contributions to the study of the fission products were made by W.H. Sullivan, A.S. Newton, and coworkers. J.C. Warf and his group made important analytical chemical contributions. I.B. Johns, Jr., H.A. Wilhelm, and the group at Iowa State worked on pyrometallurgical methods for the extraction of plutonium from uranium and for its decontamination from the fission products. Interesting methods which depended on extraction with liquid metals

and volatilization from molten uranium at high temperatures were investigated.)

Since the bismuth phosphate process did not recover the uranium for reuse, a program to find a more suitable separation process for long-range use was initiated at the Metallurgical Laboratory in 1944. Early that year Seaborg conceived of the principle of alternating between the plutonium (III) oxidation state and a higher oxidation state or states as the basis for separating plutonium from uranium and decontaminating it from fission products by the use of organic solvents. The first application of this principle was in the "Redox Process," developed at the Metallurgical Laboratory beginning in 1944 and, on the basis of much additional work there and at other sites, put into operation by the scientists and engineers of the General Electric Company at Hanford in 1951. Among the pioneer workers were W.J. Blaedel, who had the faith and perseverance necessary to override the initial difficulties, and H.H. Hyman, S.Lawroski, I.J. Schaffner, and L.R. Dawson, all of whom received much important support and encouragement from T.R. Hogness, in the face of great Manhattan District official resistance to the development of such a much-needed substitute process. In fact this important and rather large-scale effort was carried out by the group as essentially "bootleg research" during much of 1945. J.O. Maloney, J.B. Tepe, and coworkers also carried out early investigations on the semiworks scale. The redox process comprised a number of cycles in the course of which the required separations were effected. The first cycle consisted of three columns in which plutonium and uranium were separated from each other and the bulk of the fission products. The feed solution was a uranyl nitrate solution containing a minimal amount of nitric acid and heavily salted with aluminum nitrate. Oxidation with sodium dichromate converted plutonium to the solvent-extractable (VI) state, and both the uranium and the plutonium were extracted into hexone. The organic phase was scrubbed in the same column with aluminum nitrate solution to remove fission products. In a second column, the plutonium was stripped from the organic phase by reducing the plutonium to the nonextractable (III) state,

with a reducing agent (ferrous sulfamate) which did not affect the uranium. In the third column, the uranium was stripped from the hexone with dilute nitric acid. The plutonium recovered from the first cycle was further decontaminated by additional solvent extraction cycles. The uranium product from the first cycle was evaporated and likewise subjected to a second and a third solvent extraction cycle to yield uranium essentially free of fission products or plutonium, suitable for return to an isotope enrichment plant or to a nuclear reactor.

Another interesting solvent extraction process for separating and decontaminating plutonium was developed at the University of California near the end and just after the end of the war. This procedure was conceived by M. Calvin and developed with the help of J.C. Reid, H.W. Crandall, J.F. Thomas, L.L. Zebroski, and coworkers, with later contributions on an engineering scale by T. Vermeulen, T.E. Hicks, M.W. Davis, Jr., B. Rubin, and associates. In this process, plutonium (IV) was extracted from the aqueous feed with a benzene-thenoyltrifluoroacetone (TTA) solution, leaving uranium and essentially all fission products except zirconium. Traces of fission products were scrubbed from the organic phase with dilute nitric acid. Plutonium was then stripped from the organic phase by scrubbing with a reducing agent. Plutonium (III) was not extracted by TTA to a significant extent; thus the plutonium was transferred to the aqueous phase.

Any account of other contributors to the wartime work on plutonium should not end without a description of the excellent research that took place at the Chalk River Laboratory in Canada.

Research in Britain began at a very early date and centered in the Cavendish Laboratory at Cambridge University. Production of uranium metal and uranium hexafluoride was undertaken by Imperial Chemical Industries in the Liverpool area in 1941.

Because of the danger from enemy action, and also in an attempt to integrate the British and American efforts, the British research effort was moved in 1943 to a new laboratory in Montreal, where the

staff was augmented by many Canadian scientists. The project was under the joint control of the British Ministry of Supply and the Canadian National Research Council. Some members of the British research team joined laboratories in the United States, but collaboration was never very close.

The Montreal laboratory was under the direction first of H. von Halban and later of J.D. Cockcroft. E.W.R. Steacie of the Canadian National Research Council was deputy director. L. Kowarski also was a key figure in the operation of the Laboratory. The chemistry group, under the direction first of F.A. Paneth and later of J. Gueron and B. Goldschmidt, made studies of plutonium chemistry and of separation methods suitable for large-scale operation.

The chemical properties of plutonium were studied by H.G. Heal, A.G. Maddock, and B.G. Harvey, including investigation of oxidation and reduction reactions between the III, IV, and VI states. Many compounds of plutonium in these states were prepared. A survey was made of the reactions of Pu^{3+} and Pu^{4+} with a large number of organic complexing reagents. A second group, under the direction of B. Goldschmidt, studied methods for the large-scale extraction of plutonium from uranium fuel elements. L.G. Cook and R.W. Spence, who later became directors of the chemistry groups at Chalk River and Harwell respectively, were members of Goldschmidt's group. Many organic solvents were tested for suitability. The process which was finally chosen was based on the preferential extraction of Pu^{4+} from aqueous ammonium nitrate solutions by the solvent triglycoldichloride, known as the "trigly process." A plant using this process was later built at Chalk River and operated successfully.

In 1944 a large part of the Canadian effort was moved to a new permanent site at Chalk River, Ontario, about 120 miles west of Ottawa on the Ottawa River. A little later, the British group began to move back to its permanent headquarters at Harwell, near Oxford. J.D. Cockcroft went to Harwell as director, and W.B. Lewis took over the scientific direction of the Chalk River effort.

3.9. Properties of Plutonium

The successful operation of the reactor and plutonium extraction plant at Oak Ridge, Tennessee, resulted in the availability of first milligram, and then gram, amounts of plutonium early in 1944. The availability of milligram amounts of plutonium led to the immediate discovery of the III oxidation state. The early tracer work at the University of California, Berkeley, in 1941 had established the existence of a lower oxidation state (IV and/or III state) and a higher state (VI and/or higher state); and the ultramicrochemical work late in 1942 and early in 1943, as mentioned above, had defined the existence of the IV and VI states. The III oxidation state was discovered early in 1944 by R.E. Connick, W.H. McVey, and G.E. Sheline (who actually worked with about 0.25 mg of cyclotron-produced plutonium) at the University of California, Berkeley, and, essentially simultaneously, by J.C. Hindman, K.A. Kraus, J.J. Howland, Jr., and B.B. Cunningham at the Metallurgical Laboratory and D.F. Mastick and A.C. Wahl at the Los Alamos Laboratory; the latter two groups utilized the milligram amounts of plutonium made available at that time through the operation of the reactor and chemical separation plant at the Clinton Laboratories. The existence of the V oxidation state was established in the summer of 1944, through the use of plutonium obtained from the Clinton Laboratories, by Connick, M. Kasha, McVey and Sheline at the University of California, Berkeley.

As described above, the production early in 1945 of larger amounts came as a result of operation of the Hanford plant. Continuing investigations of the chemical properties of plutonium in many laboratories throughout the world, as it became available, have led to the situation where the chemistry of this relative newcomer is as well understood as is that of most of the well-studied elements. Thus, plutonium has the four oxidation states — III, IV, V, and VI — leading to a chemistry which is as complex as that of any other element. In fact, it is unique among the elements, in that these four oxidation states can all exist simultaneously in aqueous solution at

an appreciable concentration. As a metal, too, its properties are unique. It has six allotropic forms in the temperature range from room temperature to its melting point (640°C), and some of these have properties not found in any other known metal.

3.10. Publication

Accounts of the large amount of work done and results obtained on the Metallurgical Project were published in thousands of secret wartime reports. Most of these that have worthwhile scientific data in them, except those directly related to weapons, have been declassified. Their preliminary nature and the haste with which many were written make them a poor ultimate source of information. Fortunately, a good portion of the essential data was published in a more coherent fashion in various ways after the war. One of these methods of publication was the Plutonium Project Record of the National Nuclear Energy Series, published by the McGraw-Hill Book Company. R.S. Mulliken was in charge of the planning and implementation of this series, and he had the early help of L.L. Quill, E. Rabinowitch, and H.H. Goldsmith. J.C. Warner also made important contributions to the organization of this huge writing task. H.D. Young served as technical editor and then, in the summer of 1946, became general editor for the period that saw the completion of writing, declassification, and actual publication of the volumes. The overall responsibility for the publication of this record, as well as that of other volumes covering work done in connection with the Manhattan District, was centered in the Atomic Energy Commission's Technical Information Division, headed by A.F. Thompson. The volumes of especial interest from the present standpoint are Volume 14A, *The Actinide Elements*, edited by J.J. Katz and Seaborg; Volume 14B, *The Transuranium Elements: Research Papers*, edited by Katz, W.M. Manning, and Seaborg; Volume 5, *The Chemistry of Uranium*, edited by Katz and E. Rabinowitch; Volume 9, *Radiochemical Studies: The Fission Products*, edited by C.D. Coryell and N. Sugarman; Volume 17A, *Production and Separation of U²³³*, edited by L.I. Katzin

and Seaborg; and Volume 17B, *Production and Separation of U²³³, Collected Papers*, edited by Katzin. The latter two were published by the Office of Technical Services, Department of Commerce, Washington, D.C.

For a number of years following the war, some of the basic research work on plutonium was subject to rather long delay before it could be declassified. However, the situation gradually reached the point where basic research results could be published almost as soon as they were obtained, and this traditional publication procedure soon applied to all parts of the world. The Conference on the Peaceful Uses of Atomic Energy held in Geneva, Switzerland, in 1955 led to the declassification and publication in its proceedings of much information on the separation processes for plutonium and its use in reactors, and now the power technology of plutonium is handled in the traditional manner in the literature. Thus, at the present time, essentially all of the information on plutonium, except that related to its use in weapons, is being published in all countries in the traditional manner, as it is being obtained. We can, therefore, look forward to finding the continuing story of plutonium in such publications.

The plutonium story is a continuing one. The fact that the concern here has been in the main with the wartime work accounts in part for the fact that our story has covered mainly American contributions, and even among these it has been necessary within the scope of this short review to omit the mention of many people who made significant contributions. The continuing development is involving many contributions from laboratories in all parts of the world. It seems clear that this element will continue to play an important role in the history of the world.

3.11. The Franck Report

In June 1945 six committees were established at the Met Lab to make recommendations to the government regarding postwar policy. One was a Committee on Social and Political Implications. It was headed

by German-born James Franck, a venerated Nobel laureate (1925) in physics. Seaborg was made a member of this group. Other members, all chosen by Franck, were Donald Hughes, James Nickson, Eugene Rabinowitch, Joyce Stearns, and Leo Szilard. The committee's report, shaped mainly by Szilard, with some drafting help from Rabinowitch, was completed on June 11, 1945, and signed by every member of the group. It made, basically, three points. The first was that the United States could not avoid a nuclear arms race through a policy of secrecy. The second was that the best hope for national and world safety from the consequences of the bomb lay in international control of atomic energy. The third was that the military use of the bomb against Japan was "inadvisable," because it would "sacrifice public support throughout the world, precipitate the race for armaments and prejudice the possibility of reaching an international agreement on the future control of weapons." The committee suggested, instead, that the power of the bomb first be demonstrated in an uninhabited "desert or barren island."

The Franck Committee's recommendation of a demonstration rather than direct military use received very little consideration in Washington. In fact, the decision to use the bomb on two Japanese cities, without warning, appears to have been made before the Franck Report was delivered to President Truman.

3.12. Disposal of Plutonium

The intervening years of the Cold War saw the buildup of the stockpile of weapons plutonium in both the United States and the Soviet Union to the level of hundreds of tons. With the end of the Cold War, we are now faced with the dilemma of disposing of these huge quantities of plutonium. One option being considered is to mix the plutonium with fission products and then immobilize it in a glass or ceramic form for eventual placement in a geologic repository. Another option is to immobilize the plutonium by incorporating it in a mixed oxide (MOX) fuel, a mixture of plutonium and uranium oxides, and use this to fuel existing commercial power reactors.

Russia is inclined to exploit the use of weapons plutonium as fuel in nuclear power reactors and opposes the first option (immobilization) on the grounds that the plutonium could be recovered for use in nuclear weapons. The U.S. Department of Energy, in December 1996, made the decision to pursue both options, saying that the ultimate decision will depend on the results of future technology developments and demonstrations, site-specific environmental analyses, and detailed cost proposals as well as nonproliferation issues. The result of these efforts and negotiations with Russia and other nations will ultimately determine the timing and the extent to which either or both technologies are employed.

Some countries, notably Japan and France, foresee a broad use of civilian stocks of plutonium as fuel for breeder reactors or other types of advanced nuclear reactors. The quantity of such plutonium is even larger than that in military stocks and is growing much faster, by some 60–70 metric tons each year. Close to 100 tons of civilian-reactor-grade plutonium is now stored in separated form in various countries, and a much larger quantity, about 600 tons, is present in the unprocessed spent fuel from civilian power reactors.

Another legacy of the Cold War program is the negative environmental impact of the concomitant pileup of nuclear materials at the various production and fabrication sites. The cleanup programs will require the expenditure of huge sums of money over the coming decades and the training of a cadre of scientists to give direction to this effort.

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Chapter 4

Americium and Curium

The following is quoted from a talk entitled "The 25th Anniversary of the Discovery of Americium and Curium — Elements 95 and 96 — 25 Years Ago," presented by Glenn T. Seaborg at the Robert A. Welch Foundation Conference on Chemical Research, XIII, "The Transuranium Elements — The Mendeleev Centennial,"^{4.1} which was held in Houston, Texas, on November 17–19, 1969.

"We are gathered here to observe the 25th anniversary of the discovery of two very interesting synthetic elements — the transuranium elements with the atomic numbers 95 and 96. The discovery experiments for these elements, which were given the names 'americium' and 'curium,' were performed at the University of Chicago in the wartime Metallurgical Laboratory in the New Chemistry Building.

I am particularly pleased that my coworkers in these experiments, Albert Ghiorso, Ralph A. James, and Leon O. (Tom) Morgan, can be with us and participate in this program (Fig. 4.1). I am also glad to see so many others who were members of the Metallurgical Laboratory when this work took place or who have a special interest in these elements.

The discovery experiments took place during 1944 and early 1945. In the course of my remarks, I shall attempt to trace for you a sort of blow-by-blow account, including the dates when the critical observations were made. I found the preparation of this talk a most fascinating endeavor, steeped in nostalgia as I searched the deep recesses of my memory, racked the brains of my colleagues, and pored over our notebooks of that era of 25 years ago. This is in itself

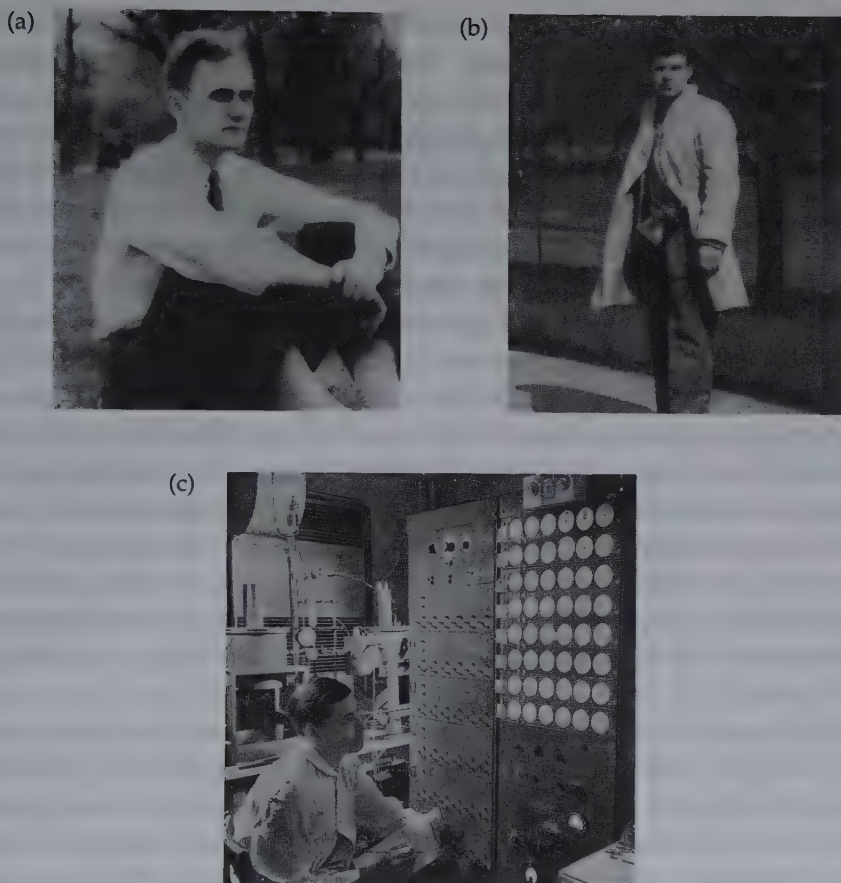


Fig. 4.1. Discoverers of americium and curium: (a) Leon O. Morgan (1944); (b) Ralph A. James at Brookfield Zoo, Chicago (1945); and (c) Albert Ghiorso in the Met Lab counting room (January 1946).

a moving experience, bringing back as it does the many emotional reactions, including heartaches and triumphs, of that time. The understanding and interpretation of the results took place over a longer period of time and had more of the ingredients of a detective story than was the case for the other elements in whose discovery I had the privilege to participate.

A number of factors contributed to the special status that this research holds in my memory. The experimental tools available at that time were so crude in relation to the deductions that had to be made. And a real breakthrough in thinking had to be made in order to devise the chemical procedures that became necessary in order to identify the new elements. I believe that I derived more personal satisfaction from these experiments than from those concerned with the discovery of the other transuranium elements.

New Chemistry, the building where this work was done, stood on the east side of Ingleside Avenue, extending south from 56th Street. It was hastily constructed during the fall of 1942, when it became apparent that the quarters on the fourth (top) floor of the George Herbert Jones Laboratory were becoming grossly inadequate for our group of chemists working as part of the Metallurgical Laboratory. I was responsible for the chemists working on the chemical processes to be used in the extraction of the plutonium from neutron-irradiated uranium and on the basic chemistry and purification of plutonium, and I, of course, was devoting my full time and energy to this task.

By December of 1943, however, so much progress had been made on these problems that I felt that I could devote part of my efforts to the synthesis and identification of the transplutonium elements with the atomic numbers 95 and 96. I asked Ralph James, a young chemist from Berkeley who had been especially proficient in the investigations of the radiochemistry of plutonium, to devote himself to this problem.

We began by attempting to produce isotopes of element 95 through bombardment of Pu^{239} with deuterons. The target material Pu^{239} was then just becoming available in milligram amounts as a result of its production in the uranium-graphite reactor at the Clinton Laboratories in Tennessee.

Ralph and I went to St. Louis at the end of January 1944, and with the help of Harry Fulbright, we bombarded 0.1-mg and 1-mg samples of Pu^{239} with the deuterons furnished by the cyclotron at Washington University. Measurements were made on the bombarded

Pu^{239} , immediately at St. Louis and later in Chicago, without any attempt to chemically separate the products. Alpha-particle range measurements were made in order to try to detect the presence of α -particle emitters of higher energy than that of Pu^{239} . These crude experiments gave negative results.

The 1 mg of irradiated Pu^{239} and two other larger samples of Pu^{239} that were bombarded with deuterons, one in the cyclotron at Washington University and the other in the 60-Inch Cyclotron in the Radiation Laboratory of the University of California, Berkeley, during the following months, were subjected to a number of chemical procedures. Chemical fractions were isolated, and their radiations examined, on the basis of several hypotheses concerning the chemical properties of element 95. These included various assumptions concerning the solubility properties of the compounds of element 95 and the potential for its oxidation from a lower state in which its fluoride is insoluble to an upper one in which its fluoride is soluble, and also included the assumption that it could not be so oxidized at all. (The elements immediately preceding element 95 — uranium, neptunium, and plutonium — each have a III and a IV oxidation state that have insoluble fluorides and a VI state that has a soluble fluoride. Increasingly strong oxidizing agents — as we go from uranium to neptunium to plutonium — are required to attain the VI state.) Unique α -particles with energies different than those of Pu^{239} were looked for and in no case were they found. In retrospect, this is not surprising because we now know that the isotopes that would have been produced, such as 95^{240} and 95^{239} , decay overwhelmingly by electron capture and to such a small extent by α -particle emission that this could not have been detected with the available techniques. These techniques did not permit detection of electron-capture decay in the presence of the tremendous amount of rare earth fission product (beta and gamma) activity.

Also beginning in January 1944, in parallel experiments, we used the reactor at the Clinton Laboratories in Tennessee to irradiate samples of Pu^{239} with neutrons. Although it was thought at that time to be unlikely, we wanted to look for the production of Pu^{240} . If such

an n,γ reaction could occur to an appreciable extent in competition with fission, then successive n,γ reactions might occur leading to a β -particle-emitting plutonium isotope and hence to an element 95 daughter. Again, in retrospect, we now know that the near identity of the α -particle decay properties of Pu^{240} and Pu^{239} made impossible the detection of the small concentration of Pu^{240} present in those neutron-irradiated samples.

An important factor in the measurement of the radiation from the isotopes of elements 95 and 96 that were being sought was the determination of the energy of α -particles by absorption or range measurements. The early absorption measurements that I shall report here were performed with thin mica absorbers and are translated into an equivalent range in terms of centimeters of air at standard conditions. Albert Ghiorso was the Group Leader of our Instruments and Physical Measurements Group and during this period he began to give more and more of his time to this problem. As the work progressed, he devised instrumentation and techniques of increasing complexity as required. Other members of his group, particularly Arthur H. Jaffey, also gave us very valuable help.

During the spring of 1944, our search also had some of the human elements dramatized by James D. Watson in his book, *The Double Helix*. Visitors from Berkeley brought the news that John W. Gofman and his group had good evidence for the identification of element 95 among the products of the neutron irradiation of Pu^{239} . I had good reason to feel that Jack Gofman might actually have beaten us to the discovery of element 95. I knew him as my brilliant graduate student whose thesis problem was the discovery and proof of the fissionability of U^{233} , a discovery that opened the use of the world's supply of thorium as a nuclear fuel. On a trip to Berkeley at the end of March, I learned the details of Jack's work. He and his coworkers had identified a 40–60-day β -particle emitter which had chemical properties similar to the rare earth elements (which was reasonable for element 95) and which was at least partially chemically separable from the rare earths. Two weeks later, after I had returned to Chicago, I received a copy of Gofman's first written description of

his work. It immediately occurred to me that his activity was actually the fission product, 57-day Y^{91} , and when I communicated this thought to him, Gofman accepted this interpretation immediately. Also, in this period of time, Joe Kennedy, during his visits to Chicago, hinted, or at least seemed to me to hint, that he, Art Wahl, and coworkers at Los Alamos had uncovered evidence for trans-plutonium elements. Again, knowing the extraordinary competence of my coworkers in the discovery of plutonium, I had more than adequate reason to believe that this might have occurred, but nothing happened to confirm this possibility.

Following these experiments performed in the first half of 1944, which gave negative results but provided much valuable experience, the first breakthrough came in July. The first bombardment of Pu^{239} with helium ions (32 MeV) took place in the Berkeley 60-Inch Cyclotron on July 8–10. By this time, the actinide concept had crystallized in my mind to the extent that we decided to proceed solely on the assumption that elements 95 and 96 could not be oxidized in aqueous acidic solution to soluble fluoride states (i.e., they should exhibit only the III or IV oxidation states that have insoluble fluorides). The sample was sent to Chicago by air and the chemical procedures started on July 12. A 2.2-mg portion of the target Pu^{239} was oxidized in aqueous solution to the plutonium (VI) oxidation state (whose fluoride is soluble) and lanthanum fluoride was precipitated. This precipitate, presumably containing the insoluble fluoride of an element 95 or 96 isotope and a small fraction of the plutonium (nearly all of which remained in solution in its soluble oxidized form), was then dissolved. Most of the small amount of remaining plutonium was oxidized and lanthanum fluoride was again precipitated to carry the element 95 or 96. The cycle was then repeated a third time. This final sample (labeled 49 α A-#9) contained only about 0.004% of the original 2.2 mg of Pu^{239} , i.e., about 0.09 micrograms or about 12,000 α -particle disintegrations per min.

On July 14, 15, and 16, careful absorption measurements in mica sheets were made on this sample. A plot of these data revealed distinctly the presence of α -particles of longer range than those of

Pu²³⁹. The original graph, a photographic reproduction taken from our notebook, is shown in Ref. 4.1, p. 227. This uses the wartime code designation '49' for the target isotope Pu²³⁹. The significance of this observation is perhaps best summarized by quoting here the entry we made in our notebook (No. 221B) on July 14, 1944:

'Plotting and comparing this data shows that a 94²³⁹ sample 20% larger than 49αA-#9 falls to 0 c/m much faster than does 49αA-#9 itself. This definitely shows the presence of a long-range α-emitting isotope in sample 49αA-#9. This is undoubtedly due to a product of the nuclear reaction of alphas on 94²³⁹ and is probably one of the following: 95²⁴² (by α,p reaction) or 96²⁴² (by an α,n reaction) or 96²⁴¹ (by an α,2n reaction). Other isotopes are possible, but these seem the most likely. The isotope 95²⁴¹ from α,n,p is fairly probable. It is difficult to estimate the range of the α-particles from this new isotope very accurately, but it would seem to be about 4.65±0.15 cm of air.'

Incidentally, the range of 4.65 cm corresponds very well with the 6.1-MeV energy of the α-particles from 96²⁴² as known today.

The sample 49αA-#9, a mixture of a few hundred disintegrations per minute of the new long-range α-activity and 12,000 disintegrations per minute of Pu²³⁹ α-activity, was then subjected to several additional oxidation cycles to further remove the Pu²³⁹. These were successful and by August 10, 1944, the sample was essentially free of Pu²³⁹. The sample was used to study the tracer chemical properties of the new isotope. Following is a verbatim extract from the Metallurgical Laboratory progress report (CS2135) covering the period of August 1944:

'About 10 mg of Pu²³⁹ were bombarded to the extent of 40 μah with 32-MeV helium ions in the Berkeley 60-Inch Cyclotron. There was found in this material a new radioactivity, emitting α-particles of range about 4.7 cm, which seems to be due to an isotope of element 96 or 95. This new radioactivity is carried quantitatively by

lanthanum fluoride, even in the presence of oxidizing agents such as dichromate or silver persulfate, indicating that this isotope cannot be oxidized to a +6 oxidation state in aqueous solution. The activity is carried by lanthanum oxalate in the presence of excess alkaline oxalate and not carried from acid solution by zirconium phenylarsonate, lead sulfate, or bismuth phosphate.'

Let me interrupt the narrative momentarily to note that as I was studying the notebooks which recorded all of these technical events, I came across the following entry, dated June 15, 1944, in Ralph James' notebook: 'Time out to get married!' Apparently, even discovering new elements wasn't allowed to interfere with love, although Ralph was back at work on June 19.

In the following months, it became increasingly apparent that this activity must be due to 96^{242} . The definitive evidence for this isotope assignment came later through the observation of this same α -activity as a product of the neutron bombardment of Pu^{239} .

As the volume of the work and the complexity of the problem increased, we felt the need for more help. Early in September I asked Tom Morgan — a young chemist who had come to us from the University of Texas and who, like James, had distinguished himself in the investigations of the radiochemistry of plutonium — to join James, Ghiorso, and me in our search. He immediately joined Ralph in performing the chemical separations on a large (200 mg) sample of plutonium which in August had received a very intensive deuterium bombardment in the St. Louis cyclotron. (Plutonium was now available in such quantities as the result of its production in the Clinton reactor.) The chemical procedure consisted of isolating a fraction presumed to contain element 95 by separating it from plutonium through repeated oxidation of plutonium to its soluble fluoride form and carrying the nonoxidizable element 95 as its insoluble fluoride on lanthanum fluoride.

There were some indications of an α -particle of range longer than that of Pu^{239} in this element 95 fraction. On the basis of Ghiorso's absorption measurements, this α -particle seemed to have a range of

about 4.0 cm in air (compared to 3.7 cm for Pu^{239}) corresponding to an energy of 5.5 MeV. Tracer experiments were carried out during September and October using a total of about 100 disintegrations per minute of this 4.0-cm α -emitter. These showed that it could be chemically separated from all the natural radioactive elements (lead and above) except actinium, thorium, and possibly protactinium. The range of the α -particles seemed to be inconsistent with their being due to isotopes of any of these elements.

In retrospect, it seems we were catching a glimpse of 95^{241} (whose α -particles have a range of 4.0 cm) produced by a d,n reaction on the tiny amount of Pu^{240} (approximately 0.01%) present in the plutonium. The observed intensity of 4.0-cm α -particles (about 100 disintegrations per minute) was consistent with the now known yield for this reaction. Another source can now also account for the observed 4.0-cm α -particles, suggesting that both sources made appreciable contributions to the observed intensity. Originally present in the plutonium (before the deuteron bombardment) was a very, very small concentration of Pu^{241} (approximately $10^{-5}\%$). As we know now, the Pu^{241} is long-lived and thus its β -particle decay would continually produce the daughter isotope 95^{241} before, during, and after the deuteron bombardment. The amount of Pu^{241} and the time of its decay are consistent with the observed intensity of 4.0-cm α -particles.

As shown in Fig. 4.2, which is a photographic reproduction of the original plot in our notebook, the presence of an α -particle emitter other than Pu^{239} could be clearly distinguished by α -particle absorption experiments with mica absorbers. In the graph, the mica absorption range for the new isotope was again converted to its equivalent air thickness — 4.05 cm. Quoting from our notebook (No. 727B) entry for October 17, 1944:

'Among the heavy isotopes ($Z=80-94$) Ac, Th, and Pa might follow the observed chemistry. This activity would have to be a new isotope of these elements, however, since none of the known ones would have this range, or behave in this way (growth, decay). Element 95 would also be expected to have this chem[istry].'

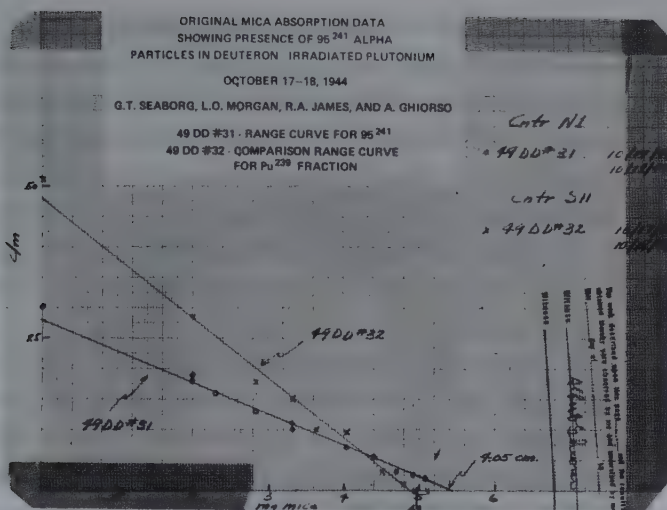


Fig. 4.2. Original mica absorption data showing the presence of 95^{241} alpha particles in deuterium-irradiated plutonium, October 17-18, 1944. G.T. Seaborg, L.O. Morgan, R.A. James, and A. Ghiorso. 49 DO #31; range curve for 95^{241} . 49 DO #32; comparison range curve for Pu^{239} fraction.

However, this activity had a somewhat will-o'-the-wisp character, and the tiny supply was finally frittered away in the course of the chemical manipulations.

It was the neutron irradiation of Pu^{239} to a relatively large total exposure, first at the Clinton Laboratories and then at the Hanford Engineer Works in the state of Washington, that led to the definite observation of an isotope of element 95 and the definite identification of the above-described product of the helium ion bombardment of Pu^{239} as an isotope of element 96.

Two samples of Pu^{239} — one of 4.4 mg and the other 8.2 mg — were placed in the reactor at the Clinton Laboratories on June 5, 1944. The first of these was removed on November 10 and returned to Chicago on November 19. This sample was put through the same type of chemical procedure described above, in which the plutonium was oxidized to the VI oxidation state whose fluoride is soluble, and insoluble lanthanum fluoride was precipitated with the intention of

carrying any nonoxidizable isotopes of elements 95 and 96 that might be present. After several such cycles had been completed, α -particle absorption measurements were made on the fraction containing any nonoxidizable components to determine the ranges of the emitted α -particles. A plot of these data on December 6, 1944, showed that, in addition to the α -particles from the Pu^{239} remaining in the sample, a component with α -particles of longer range than those of Pu^{239} was present at an intensity of a couple hundred disintegrations per minute.

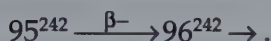
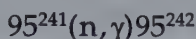
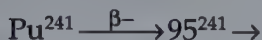
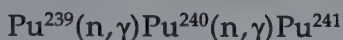
This was very exciting. Here again, as in the case of the long-range α -particles that had been observed the previous July in the nonoxidizable fraction isolated from Pu^{239} bombarded with helium ions, and during September and October in plutonium bombarded with deuterons, was evidence that radiation from an isotope (or isotopes) of element 95 or 96 had been observed. This was confirmed during the following week, when additional chemical cycles of the same type continued to remove Pu^{239} while the intensity of the long-range α -particles remained constant.

By the end of December, continuing very careful absorption measurements on the long-range α -particles indicated the possible presence of two components.

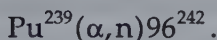
The 8.2-mg Pu^{239} sample was removed from the Clinton reactor on December 3 and returned to Chicago. This was also subjected to the now well-developed oxidation and lanthanum fluoride precipitation procedure to remove the Pu^{239} and concentrate the long-range α -particle emitter (or emitters). Again, long-range α -particles were found and in greater quantities, as would be expected on the basis of the longer neutron irradiation.

Now we were sure that we had observed transplutonium isotopes, but there remained the task of putting together all of the pieces of the puzzle. These began to fit together at about the turn of the year, early in January 1945. It was becoming increasingly clear that the long-range α -particles from the neutron-bombarded Pu^{239} consisted of two components. But it was exceedingly difficult to define the two ranges with any accuracy.

Gradually, we were able to conclude that the ranges of these two α -particles in air were about 4.0 cm and 4.7 cm (compared to 3.7 cm for Pu^{239}). Thus it appeared that the 4.0-cm α -emitter was due to 95^{241} and the 4.7-cm α -emitter was due to 96^{242} , produced in neutron-irradiated Pu^{239} by the following reactions:



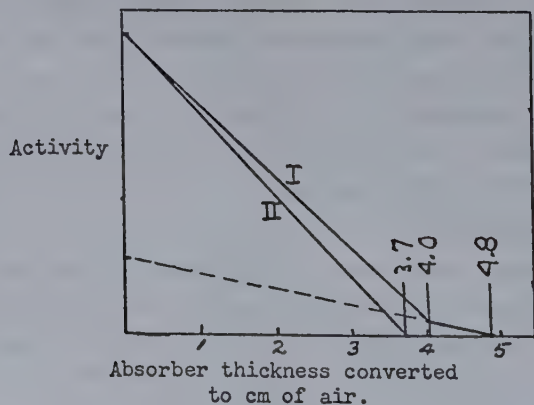
This same isotope 96^{242} was produced in helium-ion-bombarded Pu^{239} by the reaction



Aiding us in coming to this conclusion was the information we obtained by use of a 25-mg sample of Pu^{239} which had received a relatively short neutron irradiation in November in the much higher neutron flux of one of the large plutonium production reactors at the Hanford Engineer Works. This sample, which was received on January 15, 1945, was subjected to our by-now-standard chemical separation procedure. The element 95 and 96 fraction showed the same two-component long-range α -particles as had been observed in the Pu^{239} samples irradiated in the Clinton reactor. Consistent with the higher neutron flux, the yields were much larger than had been obtained in the Clinton irradiations. In fact, it was an enormous quantity by our standards, of the order of 50,000 α -disintegrations per minute. However, there were some aspects of the yield of these α -emitters that puzzled us at first. Because 96^{242} is produced in a higher order neutron absorption sequence than is the case for 95^{241} , it was expected that the ratio of the yield of 96^{242} to that of 95^{241} would be substantially higher in the high flux Hanford irradiation than in the lower flux Clinton irradiation. (The total integrated exposure was much greater at Hanford than at Clinton.) Actually

this ratio was nearly the same in the Hanford and Clinton irradiations. However, we soon understood the reason for this. The short duration of the Hanford irradiation and the relatively long interval from the end of this irradiation until we received the irradiated plutonium favored the relative buildup of 95^{241} , whereas the long duration of the Clinton irradiation allowed the 95^{241} to have a relatively longer exposure to neutrons which favored the production of 96^{242} .

These conclusions were presented at a meeting of the basic chemistry, recovery, and instrument groups of my chemical section at the Metallurgical Laboratory on January 31, 1945. Figure 4.3 is a photographic reproduction of a graph which was shown at that meeting and which was printed in the minutes of the meeting (MUC-GTS-1346, February 2, 1945). The graph synthesizes the α -particle absorption measurements made on samples chemically separated from the neutron bombardments of plutonium. This represents a clear recognition of the formation of the two isotopes 95^{241} and 96^{242} , with ranges in air for the α -particle, of 4.0 and 4.7–4.8 cm, respectively.



4.3. Summary α -particle absorption curves presented at the Metallurgical Laboratory meeting, Chicago, January 31, 1945. G.T. Seaborg, R.A. James, L.O. Morgan, and A. Ghiorso. I: Range curve for 95^{241} and 96^{242} . II: Comparison range curve for Pu^{239} .

A good summary of our understanding at that time can also be seen in the following verbatim quotation from the Metallurgical Laboratory progress report (CS-2741) covering the period of February 1945:

'Some very interesting new α -radioactivity has been found, both in plutonium irradiated with neutrons in the Clinton pile and plutonium irradiated with neutrons in the Hanford pile. This α -activity exhibits just the sort of chemical behavior which has been predicted for the transplutonium elements. For example, it is carried by rare earth fluorides and it has not yet been possible to oxidize it to a state or states where its fluoride is soluble. It seems to be chemically separable from all the rest of the 94 elements, and the best present interpretation is that it is due to element 95 and/or element 96. The α -activity is composed of two components, one of range 4.0 cm and the other of range 4.7 cm. A very attractive possibility is that the 4.0-cm α -activity corresponds to 95^{241} formed from the β -decay of 94^{241} which comes from the reaction $94^{240} (n,\gamma) 94^{241}$, and the 4.7-cm α -activity corresponds to 96^{242} (cf. reports CS-2124 and CS-2135) from β -decaying 95^{242} following the reaction $95^{241} (n,\gamma) 95^{242}$. The ratio of the yields in the Hanford as compared to the Clinton bombardment seems to be proportional to the second power of the total neutron irradiation, as would be expected on the basis of these particular isotopic assignments, although the accuracy of the estimation of the neutron fluxes is not sufficient to make this at all certain.'

Soon after our results had been communicated to scientists at Los Alamos, they made a mass spectrographic identification in purified plutonium of a relatively volatile isotope with the mass number 241, which they identified with our 95^{241} . Since this could be extracted by volatilization from pure plutonium (from which element 95, which was originally present, had been removed in the purification process), we knew that the parent isotope, Pu^{241} , must have a relatively

long half-life (of the order of months or longer). Our own work had not given us any information concerning the half-life of Pu^{241} . In order to test for the presence of Pu^{241} , we subjected 250 mg of purified plutonium, which had been produced in a Hanford reactor, to our chemical procedure for the isolation of element 95. By March 20, we had a purified fraction containing the extracted 95^{241} . The intensity of the 4.0-cm α -particles corresponded to about 30,000 disintegrations per minute. This established beyond doubt the identity of our 4.0-cm α -emitter. Also, we now had a large sample with which to study the chemical properties of element 95 and, beyond that, we knew that we had an inexhaustible supply in our large stock of plutonium. Subsequent 'milkings' led to continued extractions of the 4.0-cm α -emitter from plutonium samples and we were able to establish the half-life of Pu^{241} (now known to be 13 years).

Also in March, we had another bombardment of Pu^{239} with helium ions in the Berkeley 60-Inch Cyclotron. Again, we chemically separated an element 96 fraction using our well-established procedures. Imagine, then, our surprise and consternation when the α -particles emitted in this fraction had a range of 5.0 cm rather than 4.7 cm of the expected 96^{242} . However, we were able soon to solve this additional puzzle. It happened that the Berkeley cyclotron had been modified since our July bombardment so that it now delivered 40-MeV helium ions rather than the previous 32-MeV energy. This meant that isotopes such as 96^{241} and 96^{240} would now be produced in predominating yields, due to $\alpha, 2n$ and $\alpha, 3n$ reactions, in addition to the previously produced 96^{242} . It was possible later to assign the 5.0-cm α -emitter to the new isotope 96^{240} and to detect the lower intensity α -particles due to the 96^{242} .

A few months later, in July, the isotope Pu^{238} was identified as the α -particle decay product of 96^{242} , thus lending further confirmation to this isotopic assignment. Decay measurements showed that 96^{242} had a half-life of about five months (and 96^{240} a half-life of about one month) while 95^{241} exhibited no measurable decay (now known to have a half-life of 434 years). As soon as a sufficient quantity of 95^{241} was available as a decay product of Pu^{241} , it was irradiated with

reactor neutrons to confirm the production of 96^{242} by the reactions $95^{241}(n,\gamma)95^{242} \xrightarrow{\beta^-} 96^{242}$. Soon thereafter W.M. Manning and L.B. Asprey measured the half-life of 95^{242} as about 16 hours.

Of special interest is the fact that we were able to produce and definitely identify the isotope 95^{241} also by charged particle bombardments. Using helium ions from the Berkeley cyclotron to bombard uranium, we were able to identify Pu^{241} and its β -decay daughter 95^{241} produced by the reactions $U^{238}(\alpha,n)Pu^{241} \xrightarrow{\beta^-} 95^{241}$. The significance of this lies in the fact that we were able to obtain declassification of this method of production much before the original method for the production of 95^{241} (via the neutron irradiation of Pu^{239}) could be declassified. Therefore, my first announcement at a scientific meeting of the discovery of elements 95 and 96, in a paper presented at an American Chemical Society symposium at Northwestern University on Friday, November 16, 1945 (published in *Chemical and Engineering News*, Vol. 23, p. 2190, Dec. 10, 1945), described the production of element 95 by helium ions rather than by neutron bombardment.

As it actually turned out, the discovery of elements 95 and 96 was announced to the world for the first time prior to the symposium. This occurred on the radio program *Quiz Kids* on Sunday, November 11, Armistice Day of 1945 (Fig. 4.4). I happened to be serving as a guest on this radio program and one of the kids asked me if any new chemical elements had been discovered at the Metallurgical Laboratory during the war. As the information had already been declassified for the symposium to be held the following Friday, I replied in the affirmative to his questions. Following is a verbatim transcription of this question and answer:

Richard: Oh, another thing — have there been any other new elements discovered, like plutonium and neptunium?

Seaborg: Oh yes, Dick. Recently there have been two new elements discovered — elements with atomic numbers 95 and 96 out at the Metallurgical Laboratory here in Chicago. So now you'll have to tell your teachers to change the 92 elements in your schoolbook to 96 elements.'



4.4. "Quiz Kids" Sheila Conlan and Robert Burke with Glenn T. Seaborg when he informally announced the discovery of elements 95 and 96 on a radio show in 1945. 11/11/45.

Soon after this, on December 15, 1945, I made a guest appearance on the Watson Davis program *Adventures in Science*, which was a national network radio program at that time. Following is a verbatim transcript of a portion of that radio program:

'Announcer: By the way, I'd like to know whether you have named these two new elements that you have discovered.

Seaborg: Well, naming one of the fundamental substances of the universe is, of course, something that should be done only after careful thought. We have been faced with considerable difficulty in these cases because we have run out of planets. Naming neptunium after the planet Neptune, and plutonium after the planet Pluto, was rather logical. But so far the astronomers haven't discovered any planets beyond Pluto. So we'll have to go to some other method of naming.

Announcer: What's that, Dr. Seaborg?

Seaborg: This hasn't been decided yet. One possibility might be to rely on some property of these elements. We do have an idea for the naming of element 95 along these lines and

may have a suggestion to offer pretty soon. And, by the way, you may be interested to know that we have received lots of suggestions. Some good and some not so good.

Announcer: Well, Dr. Seaborg, perhaps some of the listeners to *Adventures in Science* will want to make suggestions as to the naming of the new elements. Will you be willing to have them write in their suggestions?

Seaborg: Well, I don't promise to follow the suggestions. But it might be interesting to know what the public thinks about naming new chemical elements.

Announcer: Very well, Dr. Seaborg. If you want to suggest names for new elements 95 and 96, just drop a postcard to Watson Davis, Science Service, Washington 6, D.C. And to all those who write in, Mr. Davis will send them a free copy of the current issue of *Chemistry* magazine, which contains Dr. Seaborg's full technical paper and a new arrangement of the chemical periodic table. It's free for the asking. So be sure to ask for the elements 95 and 96 article and address Watson Davis, Science Service, Washington 6, D.C.' (See Note at end of chapter.)

Actually, we gave a great deal of thought to the naming of elements 95 and 96. My theory that they should be chemically similar to the rare earth elements was being borne out to such an extent that we were finding it almost impossible to chemically separate them from these elements. Although we eventually succeeded, during the period of our futile efforts to do so Tom Morgan continually referred to elements 95 and 96 as 'pandemonium' and 'delirium.'

Names were finally suggested for elements 95 and 96 in the course of a talk that I gave at the annual spring meeting of the American Chemical Society in Atlantic City on April 10, 1946 (published in *Chemical and Engineering News*, Vol. 24, p. 1192, May 10, 1946). Element 95 was given the name 'americium' (symbol Am), after the Americas, in analogy to the naming of its rare earth homolog,

europium, after Europe; and for element 96 we suggested the name 'Curium' (symbol Cm), after Pierre and Marie Curie, in analogy to the naming of its homolog, gadolinium, after Johan Gadolin.

This, then, is the story of the discovery of the elements with the atomic numbers 95 and 96. I hope that I have succeeded in recapturing for you some of the excitements, frustrations, and satisfactions that we experienced in the course of this scientific adventure.

This brings me to the end of my account. I have tried to leave enough time for my coworkers to step to the podium and make any remarks that they would like to. I don't have any idea at all as to what they are going to say, but I would like to call on Albert Ghiorso first.

* * * * *

Dr. Albert Ghiorso (speaker), Lawrence Radiation Laboratory, University of California, Berkeley: I happened to glance up at one of the slides Glenn was using and I saw my name there. If you would turn your head to the side, you would see it read, 'Observed and understood by Albert Ghiorso.' I am sure I observed it, because I did most of the counting in those days. But I am sure I didn't really understand it.

Let me illustrate my point. In the early days — I think this was one of the early experiments — Ralph and I traveled down to the great City of St. Louis and we put a target on the St. Louis cyclotron, about 10 mg of plutonium. He had carefully prepared this. The idea was we would bombard it in the deuteron beam for a while and then remove it and put a piece of mica over it so that when it was placed inside a nitrogen-filled alpha chamber we could look for long range alphas from some new activity. We did that, but the amazing thing about it is that we had no hood — no survey meters — the thing must have been as hot as a firecracker. But I guess Ralph and I are still here, so we must have survived somehow.

The other story is a pretty good joke on me. After we had found 95 and 96, we had gotten larger samples of plutonium back from the Clinton pile and we were looking for new isotopes. Since we knew

we had Pu^{241} the important thing was to look for Pu^{242} . We got a beautiful sample of plutonium about a milligram per cm^2 thick — a very beautiful sample on a big 3-inch-diameter plate, carefully placed there with cement. I put this into the fission counter and we proceeded to watch for big kicks — spontaneous fission events. Time went by and after about 15 min there was an event. It was terrific. And I, of course, went dashing down the hall to tell people. About another 15 min later there was another. And then another. The Pu^{242} spontaneous fission half-life must be very short — we decided it was very exciting! It really was, for several hours. Then I happened to notice a strange thing. You know, those things are just like a train. They seem to be coming in on time. So, I took the opportunity to time them. Sure enough, every 15 min we would get a count. A look in the scope showed great big kicks, clearly fission counts, of course. Then I got an idea. I said, 'Do you suppose that it could be that the intense alpha activity is charging up the plate on its insulated backing and discharging after 15 min, giving the big pulse?' Of course, that is what it was. For a few hours we had a beautiful experiment and an unusual discovery. Perhaps ever since then I have had it in for spontaneous fission. Thank you.

Dr. Seaborg: I didn't see how he could make a rationale for that last remark on this program.

Ralph, would you like to reminisce a moment? If you remember anything about those Met Lab days.

Dr. Ralph A. James (Discussion Leader), Lawrence Radiation Laboratory, University of California, Livermore: Well, my memory of those days is very hazy, to say the least. I think that Dr. Seaborg has done a wonderful job of looking through these unintelligible notebooks and coming up with a reasonable story. I am sure he is right, but I just can't quite believe that I got four whole days off to get married.

Now, just so I won't steal any of Tom Morgan's thunder and not knowing what he is going to say either, I thought I would just say a little bit that's sort of indirectly related to the discovery of these elements.

When I first came to the Met Lab in the Spring of 1942, my very first assignment was to see if there was any possibility of oxidizing plutonium and volatilizing it with osmium. I spent many days, many weeks, using all kinds of oxidizing agents and media of various boiling points to try to volatilize the plutonium. Well, of course, it didn't work. But I think this was one of the early indications that there was something wrong with the periodic table as it existed at that time. And I think this experiment and others similar to it were directly responsible for the actinide hypothesis, which it was called in those days, and which was very directly concerned with the discovery of americium and curium.

Now, those days were certainly very hectic, as has been indicated, but they were very exciting and rewarding to me. Thank you.

Dr. Seaborg: Thank you very much, Ralph. It is with some hesitation now, after that, that I call on Tom Morgan. Tom, do you want to come forward?

Dr. L.O. Morgan (Discussion Leader), The University of Texas at Austin: What's your hesitation? I still think 'delirium' and 'pandemonium' would be better names than anything else. I don't think I had fully realized until today how much my involvement in this project depended on Ralph getting married. Somebody had to carry on the work; I was there and available.

Glenn has used some euphemisms in discussing the work that went on in those days. When he refers to an oxidation-reduction cycle, or a series of them, I don't think he fully realizes what actually went on in the laboratory when he wasn't watching. When he uses the words 'frittered away,' I am not quite sure that he is conscious of how much trauma was involved in those losses of such precious material. Another thing that he referred to was the involvement of Albert Ghiorso as a fine instrumental scientist. Again, I think that he is guilty of euphemism. One of the biggest handicaps that we had to operate under was Albert's great desire to improve his instrumentation. There are a lot of you here tonight who have participated in the work on isotopes. You have worked with Al and I think a very brief

story might be in order. We had gone through some range measurements very, very carefully on highly purified samples. I went into the counting room at one o'clock in the morning, ready to run some more samples in the range chamber. There I discovered the parts spread out over the table. I ran to the telephone and called Al to ask him what was going on and he said he was improving the instrumentation. That was a constant urge with him. If he does the same thing to the linear accelerator that he did to those nitrogen chambers, I wonder that they get out anything today.

Glenn mentioned one sample which contained a hundred counts per minute of americium. It has been one of my proud boasts that during all of our work we did a tremendous amount of chemistry on a few hundred counts per minute. After listening to some of the discussions today and having heard some of the previous ones in which they worked with 2 counts or 15 counts per minute, maybe I shouldn't be quite so proud of that. I do have a fond recollection of the instrumentation that we had in those days and when I visit the Laboratory today and see what is there, I am very envious.

I hope that the future of transuranium chemistry is as bright as it seemed to us in those days. Thank you.

Dr. Seaborg: Thank you very much, Tom.

We have time for a little rebuttal — Al, do you want to describe the instrumentation a little bit, to give some of these newcomers a little feeling for what it was necessary to work with in those days?

Dr. Ghiorso: To be honest, Glenn, I don't really remember very much about those days. I am in the same position as Ralph. It seemed as if it was a succession of 80-hour weeks — it was in Chicago — they had the hottest summers they ever had — the coldest winters — we had babies — we had golf matches.

We used to try to get off occasionally after work and go down to Jackson Park to play golf. At first I never was able to beat Glenn, although I thought I was much better than he was — until I learned the secret. He just always used the 'Jack Rivers' technique on me. If you know the story about 'Jack Rivers,' you understand Seaborg.

When I discovered his secret I had conquered it; then I knew what to do and was able to beat him a few times.

The instrumentation *was* kind of crude in those days. It really was. I pride myself on working out the method of using mica absorption curves to measure α -particle ranges at high geometry to obtain a fair amount of information. But it was really difficult and time-consuming. We took many, many counts. The mica absorbers would tend to charge up and then break down, thus giving spurious electrical counts so that we would have to do special things with them. But we somehow managed to struggle along and get the important information. Well, I guess, Ralph, it must be right — at least, we found the same isotopes later!

I think only Glenn really had the necessary confidence in this work. He was our leader and certainly deserved this appellation. I would do experiments, what were complicated experiments at that time, and do them very carefully and report back to Glenn what had happened. Perhaps, three months later, he would ask me, 'Now, what happened on such-and-such an experiment where you found this many counts with this range under these conditions?' Sometimes I found to my amazement that he remembered more about my work at that time than I did. I was always impressed because at the same time he also had the tremendous job of handling a hundred scientists, doing many complicated chemical tests in the Met Lab. I am very proud of the fact that I learned to be a scientist under Glenn's tutelage; it was really hard going, but well worth it.

Since that time, of course, things have changed a lot. If you heard my talk this afternoon, you know that one can do an awful lot with very little. But you still have to have faith, you have to have a certain amount of luck, and you have to be careful.

Those pioneer days, fortunately, are gone forever. I am sure many of you have equally gross stories about how you did science in the old days. It worked, but it was a lot of work. It is instructive to read back and see how Rutherford and his colleagues did their basic experiments with very little activity but very sophisticated ideas in their heads; their methods were often extremely crude. The work

with americium and curium was of the same genre and I think Glenn deserves a tremendous amount of credit for successfully carrying through the discoveries at this time. It took a Seaborg to do it.

Dr. Seaborg: Thank you very much. I am glad that we had the opportunity on this occasion of the Mendeleev Centennial here in Houston to have this little observance of the 25th anniversary of the discovery of these two elements with the atomic numbers 95 and 96, americium and curium.

One reason I wanted to do this at this time is that I thought that on the next occasion, which would probably be the 50th anniversary, some of us might have a little trouble recalling what had happened 50 years ago.

So, with that, thank you very much for being with us this evening."

Note

Dr. Seaborg's response to the Watson Davis radio show *Adventures in Science* on December 15, 1945:

My wife Helen and I, a month later, took time to survey the suggestions. George G. Abel, III, of Media, Pennsylvania, suggested "proxogravum" for element 95 (from the Latin *proximum gravissimus*, meaning "next-heaviest") and "gravum" for element 96 (from the Latin *gravissimus*, meaning "heaviest"). James McManus of Buffalo, New York, also suggested Latin-derived names — "alium" from the Latin *alius*, meaning "another," for element 95; and "novium" from the Latin for "new," for element 96. J.D. Boon of the Department of Physics of Southern Methodist University offered the following: 95 — "pentonium"; 96 — "sextonium"; 97 — "septonium"; 98 — "octonium"; 99 — "novanium"; and 100 — "centurium." Gertrude Strother of Ware Shoals, South Carolina, offered "quintium" for element 95 and "sexium" for element 96.

A number of people proposed naming the new elements after our sun and moon. Marjorie Salsbury of Patterson, Iowa, also wanted one of the elements to be named something that suggests the sun,

such as "solonium." Since we have used up planets, B.H. Bedell of Toronto believed we should finish off with the sun and moon and call the elements "solium" and "lunium." "Sunonium" and "moononium" were the suggestions of Miss Lillian Cousey of St. Petersburg, Florida. (Actually, an element had already been named after the sun — element 2, "helium," which had been named by the English astronomer Sir Norman Lockyer in 1868.)

Astronomical bodies were popular suggestions as still others wrote proposing names fashioned after planets, stars, constellations, or even the signs of the zodiac. Otto I. Bergh of Miami suggested "nebulium" and "solium." Arthur Nowicki of Buffalo, New York, who also submitted "sunian" since we had run out of planets, suggested "big dipperain" or "big bearianen." He was echoed by Mrs. J. Pfeiffer of Ocean City, New Jersey, who offered "dipperium" and "cometium." V.T. Johnson of Springfield, Vermont, suggested that 95 be called "stellanium" and 96 "astronium." Robert G. Harris, a high school student from South Portland, Maine, proposed that 95 be called "bolidium" and 96 "asteroidium." Hugo Bass from New York City suggested the following for elements 95 onward: "transneptunium," "siderium," "stellium," "astralium," and "cosmium."

Myron Stein of Brooklyn, New York, suggested "draconium" for element 95 after the constellation Draco and "leonite" for element 96 after the constellation Leo. Still later, George A. Davis, a lawyer, of Buffalo, New York, suggested "sirium" and "canopium" after the brightest stars. Carl Rees of Weston, Ontario, suggested the elements be named after the two moons of the planet Mars — Deimos, meaning "dread," and Phobos, meaning "terror." Karl Hirschfeld of Philadelphia suggested number 96 be named "universum." Frank C. Banham, Jr., of Kinberton, Pennsylvania, suggested "virgorium" and "ariesium" after two zodiac signs.

W.B. Barrows of Farmington, Connecticut, noted that we have not named anything after the planet earth — "terrium." He also advised that a number of the elements have been named for countries and proposed "amerium." Element 96 could be called "finium" or "ultimum."

Some thought that the elements should be named in honor of the way they were created. Walter Schwandt of Great Kills, New York, believed one of the elements should be called "artifium" or "artificial." Seymour Gottlieb of Long Island City, New York, also proposed that the elements be named after the elements they resemble with the prefix "cyclo," because they were discovered in the cyclotron. Aram Amirian of Jersey City, New Jersey, believed element 95 should be called "mechanicium" and element 96 "sciencium." Ralph O. Payne said that since 95 and 96 were discovered by the use of α -particles, element 95 should be named "alphonium." He suggested 96 could be called "cosmonium" after cosmic rays, "which will give, in the future, rise to other elements." Michael Flax of Brooklyn, New York, proposed "neutronium" and "alphanium," since the elements were produced with the aid of α -particles and neutrons.

Ernest H. Kalmus of New York wrote a long letter, saying we should go back to the very roots of the discovery and name the new elements according to the way they were found, namely by splitting the atom and fission. Therefore this would mean calling them "splittium" and "fissium." Kalmus also suggested that perhaps they could be named after the great scientists who shared in their discovery: "fermium" and "bohrium."

Other listeners also wanted to honor famous scientists. Eric R. Miller of Winter Park, Florida, suggested the names of [Henri] Becquerel and [Lord Ernest] Rutherford as men to be honored. Anthony Saletan of New York City (Manhattan) suggested "curium" for element 95 and "einsteinium" for element 96. Samuel Ross of Miami Beach suggested "einsteinium" and "rooseveltium" for the newly discovered elements.

Martin Schachne of Brooklyn, New York, believed that the elements should be named after U.S. presidents, such as "washingtonium." Seymour Gottlieb wondered if possibly one should be named after President [Franklin D.] Roosevelt and called "roosium," since he made the project possible. Samuel B. Katz of Brooklyn, New York, also

proposed the name "rooseveltium," symbol Fdr, for the man responsible for the great impetus to research in nuclear physics.

W.B. Barrows made several contributions of mythological characters, such as Vulcan, Hercules, Zeus, Venus, Apollo, and Mars.

M. Miller of the Bronx, New York, suggested for element 95 "unonium" in honor of the UNO (United Nations Organization), and for element 96 "paximum," from the Latin word for "peace." Elihu Schimmel of Lawrence, New York, said if we could not find better names, then perhaps we could use "mondium," for "worldliness," and "eternium," from "eternity."

Robert Elenko of Brooklyn, New York, liked the name "futorium" (symbol Fu) for element 96.

Warren C. Chapman of Atlantic City, New Jersey, suggested naming element 95 "seaburnium," "nutronium," or "nonagintium." Donald Steward of Kenmore, New York, suggested "xtinium" and "ytumium" for elements 95 and 96.

Irwin Alemanoff of Brooklyn, New York, said, "Since so much work on elements 93-96 has been done in the University of California, I think that it might be fitting to name element 95 'unicalium' in honor of the University. As for 96 I think we should honor the family that had so much to do with the pioneer work in radioactivity and nuclear transformations, by calling it 'curium' for the [Marie and Pierre] Curies."

A little later, Burke Brown of Guelph, Ontario, suggested the names "seadium" and "boardium" in my honor as one of the discoverers.

Another facetious suggestion for element 95 was "persephonium," after Persephone, goddess friend of Pluto. Since Pluto had kidnapped Persephone, there might well have been an offspring, so the suggestion was made that element 96 be named "bastardium."

Actually the names chosen for elements 95 and 96 and some of the even heavier elements as will be discussed in the following chapters, were encompassed (or approximated) on this fascinating list. However, the names for elements 95 and 96 were chosen in recognition of their new position on the periodic table.

At a meeting of the Heavy Isotopes Group at the Metallurgical Laboratory on March 5, 1946, I suggested that 95 and 96 be named "americium" and "curium" by analogy to the naming of their lanthanide homologs "europium" and "gadolinium." It was also pointed out that the +2 state of element 96 would be "cur-ious," but I replied that this oxidation state was not expected to exist.

Americium was first isolated by B.B. Cunningham in the form of a pure compound, the hydroxide, in the fall of 1945 at the wartime Metallurgical Laboratory; a photograph of his hydroxide is shown in Fig. 4.5. It can be prepared in gram and larger amounts by the neutron bombardment of plutonium according to the reactions shown above, and it has thus been possible to investigate its chemical properties through the use of macroscopic quantities.

Curium was first isolated in the form of a pure compound, the hydroxide, of ^{242}Cm by L.B. Werner and I. Perlman at the University of California during the fall of 1947. A photograph of this first



Fig. 4.5. First photograph of an americium compound. The hydroxide is shown in the bottom of the tube (center). Americium was first isolated in January 1945. Eye of sewing needle indicates magnification of about 15-fold.



Fig. 4.6. Photograph of the first curium compound (hydroxide) in the bottom of the microcentrifuge cone. Isolated in the fall of 1947. Magnified about threefold.

PERIODIC TABLE SHOWING HEAVY ELEMENTS AS MEMBERS
OF AN ACTINIDE SERIES

Arrangement by Glenn T. Seaborg, 1945

1 H 1.008																	1 H 1.008	2 He 4.003													
3 Li 6.940	4 Be 9.02																	5 B 10.82	6 C 12.010	7 N 14.008	8 O 16.000	9 F 19.00	10 Ne 20.183								
11 Na 22.997	12 Mg 24.32	13 Al 26.97																	13 Si 28.06	14 P 30.98	15 S 32.06	16 Cl 35.457	17 Ar 39.944								
19 K 39.096	20 Ca 40.08	21 Sc 45.10	22 Ti 47.90	23 V 50.95	24 Cr 52.01	25 Mn 54.93	26 Fe 55.85	27 Co 58.94	28 Ni 58.69	29 Cu 63.57	30 Zn 65.38	31 Ga 69.72	32 Ge 72.60	33 As 74.91	34 Se 78.96	35 Br 79.916	36 Kr 83.7														
37 Rb 85.48	38 Sr 87.63	39 Y 88.92	40 Zr 91.22	41 Nb 92.91	42 Mo 95.95	43 Tc 101.7	44 Ru 101.7	45 Rh 102.91	46 Pd 106.7	47 Ag 107.880	48 Cd 112.41	49 In 114.76	50 Sn 118.70	51 Sb 121.76	52 Te 127.61	53 I 126.92	54 Xe 131.3														
55 Cs 132.91	56 Ba 137.36	57 La 138.92	58 Ce 140.12	59 Pr 140.92	60 Nd 144.27	61 Pm 144.91	62 Sm 150.43	63 Eu 152.0	64 Gd 156.9	65 Tb 159.2	66 Dy 162.46	67 Ho 164.93	68 Er 167.2	69 Tm 168.93	70 Yb 173.04	71 Lu 174.97	72 Hf 178.49	73 Ta 180.95	74 W 183.85	75 Re 186.21	76 Os 190.2	77 Ir 192.22	78 Pt 195.08	79 Au 196.967	80 Hg 200.59	81 Tl 204.38	82 Pb 207.2	83 Bi 208.98	84 Po 209	85 At 210	86 Rn 222
87 Fr 223	88 Ra 226	89 Ac 227	90 Th 232.04	91 Pa 231	92 U 238.03	93 Np 237	94 Pu 244	95 Am 243	96 Cm 247																						

LANTHANIDE SERIES	57 La 138.92	58 Ce 140.12	59 Pr 140.92	60 Nd 144.27	61 Pm 144.91	62 Sm 150.43	63 Eu 152.0	64 Gd 156.9	65 Tb 159.2	66 Dy 162.46	67 Ho 164.93	68 Er 167.2	69 Tm 168.93	70 Yb 173.04	71 Lu 174.97
ACTINIDE SERIES	89 Ac 227	90 Th 232.04	91 Pa 231	92 U 238.03	93 Np 237	94 Pu 244	95 Am 243	96 Cm 247							

XBL 769-10603

Fig. 4.7. Periodic table of 1944 and 1945 showing Seaborg's placement of the heaviest elements as an "actinide series."

hydroxide is shown in Fig. 4.6. The isotope ^{242}Cm is so highly radioactive, because of its short half life, that chemical investigations with it in macroscopic concentrations are very difficult. Therefore, the subsequent work using weighable amounts has been done with longer-lived heavier isotopes of curium.

The article in *Chemical and Engineering News*^{4,2} in 1945 included Seaborg's 1944 revised periodic table, which placed the newly discovered transuranium elements as an actinide series (Fig. 4.7). This placement of the actinide elements, and consequent placement of the transactinide elements, has been universally accepted as represented by the current periodic table, shown in Fig. 1.5.

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Chapter 5

Berkelium and Californium

5.1. Introduction

The discoveries of berkelium, element 97, and californium, element 98, seem usually to be discussed together, partly because they were discovered in late 1949 and early 1950 and both discoveries were published in the *Physical Review* in 1950,^{5.1, 5.2} and partly because they both depended on obtaining sufficient quantities of the americium and curium needed for producing them in helium bombardments. As recounted later, this took several years to accomplish. Berkelium was produced via the $^{241}\text{Am}(\alpha, 2n)^{243}\text{97}$ reaction in late December 1949 and californium was produced in February 1950 via the $^{242}\text{Cm}(\alpha, n)^{245}\text{98}$ reaction at the Berkeley 60-Inch Cyclotron. The key to the discovery of berkelium was removal of the 7 mg of ^{241}Am target material by oxidizing it to the hexapositive state. After considerable additional chemistry, the berkelium fraction was ultimately eluted from a cation exchange column in the expected position, just ahead of curium. Californium was also separated and identified based on its expected elution position ahead of elements 96 and 97. The name "berkelium," symbol Bk, after the city of Berkeley, California, was quickly selected for element 97, by analogy to its chemical homolog, terbium, which is named after the Swedish town of Ytterby. After considerable discussion among the discoverers, the name "californium," symbol Cf, after the state of California, was chosen for element 98 even though this is not analogous to the name of its chemical homolog, dysprosium, which was taken from the Greek word *dysprositos*, meaning "hard to get at."

5.2. Reminiscences on the Discovery of Berkelium and Californium

A symposium commemorating the 25th anniversary of these discoveries was held at the Lawrence Berkeley Laboratory on January 20, 1975. All of the discoverers were there and presented their reminiscences. In addition, other speakers reviewed the research performed on berkelium and californium during the quarter of a century since their discoveries, including their nuclear and chemical properties, their large-scale production, and potential practical applications. The four reminiscences in the words of the discoverers taken from the published proceedings^{5,3} of that symposium are given here.

5.2.1. Glenn T. Seaborg

"The discovery experiments were performed up in the old Building 5 (at LBL), which still exists but is much changed today and hardly recognizable to those of us who remember its layout of a quarter of a century ago. The space was cramped, and the equipment, by today's standards, was crude, but I think we never enjoyed the scientific quest more than we did in those days.

Fortunately, we have all of the discoverers with us today. However, in talking to Stan Thompson, Al Ghiorso, and Ken Street, with the hope of going through their old notebooks with them in preparation for today, I found they have lost them. All of their notebooks, or scraps of paper with data on them, or whatever, have disappeared. And they can't find any of the equipment used for these historic experiments. What a sense of history! But of course in those days the experiment of the day was paramount, then on to the next experiment with its new equipment and new ideas. Why worry about the past?

Actually, the situation isn't as bad as I have depicted it. We have good notes covering our research meetings, much better than those we keep today (when we don't keep any), and the journal publications describing this work are unusually detailed and informative.

And we have our memories, which have not failed yet — at least not completely. Preparing for today's session has been, for me, a pleasant, nostalgic journey into the past.

I recall that we began planning for the possible synthesis and identification of transuranium elements as soon as, or even before, we returned to Berkeley from the Chicago Metallurgical Laboratory, i.e. in late 1945 and in 1946. I thought that this would be a good Ph.D. thesis problem for Stan Thompson (Fig. 5.1) and it was, of course, natural that Al Ghiorso (Fig. 5.2) would participate on the radiation detection end of the problem, as he had in the discovery of americium and curium in Chicago a year or two earlier.

On the basis of our confidence in the actinide concept we felt we could make the chemical identification, although we knew we would have to develop better chemical separation methods than were then available to us. And it seemed clear that we would use helium ion

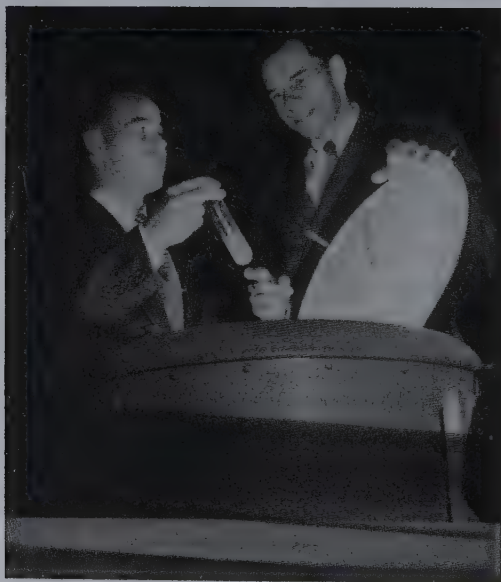


Fig. 5.1. Stanley Thompson and Glenn Seaborg in 1948, when the work on the discovery of elements 97 and 98 was in progress.



Fig. 5.2. Albert Ghiorso inserting a sample into the equipment used in the discovery of berkelium and californium.

bombardments of americium and curium for our production reactions once these elements became available in sufficient quantity through production by prolonged neutron bombardment of plutonium, and we learned how to handle safely their intense radioactivity.

We knew these things but we didn't anticipate how long it would take to solve these simple problems. Actually, three years went by before we found ourselves ready to make our first realistic experiment. Stan, who couldn't wait so long, went on and finished his Ph.D. thesis on another problem. The three years were consumed in arranging and waiting for the production of the quantities of americium and curium needed for use as target material, and developing the required extremely efficient chemical separation methods based on ion exchange and the required methods for safe handling of the intensely radioactive target material.

During the fall of 1949 we made a number of bombardments of americium with helium ions in the 60-Inch Cyclotron, with emphasis on looking for α -particle-emitting isotopes of element 97, all with negative results. It was becoming clear that we should look for electron capture (EC) decay by detecting the accompanying conversion electrons and X-rays, so Ghiorso worked to improve the detection efficiency for such radiations.

The first successful experiment was performed on Monday, December 19, 1949. A target containing 7 mg of ^{241}Am was bombarded with helium ions in the 60-Inch Cyclotron, after which the chemical separation was started at 10 a.m. After removal of the bulk of the americium by two oxidation cycles (utilizing oxidation to the hexapositive, fluoride-soluble, oxidation state of americium, which had just been discovered by Asprey, Stephanou, and Penneman at Los Alamos), the 97, Cm and remaining Am were carried on lanthanum fluoride, dissolved and subjected to a group separation from fission product lanthanide elements (using a method of elution with concentrated HCl, just discovered by Ken Street), after which the actinide fraction was put through a cation exchange adsorption-elution procedure; this entire process was completed in seven hours. The prediction that element 97 would elute ahead of Cm and Am, in sequence, was of course the key to its successful chemical identification. Figure 5.3 shows a reproduction of the data obtained that afternoon. In this case, and especially in considering the data from following elution experiments, we were somewhat surprised to see the rather large gap between 97 and curium; we shouldn't have been surprised, because there is a notably large gap between the elution peaks of the homologous lanthanide elements terbium and gadolinium.

Detected in the samples that eluted at the peak corresponding to element 97 were conversion electrons, X-rays of energy corresponding to decay by electron capture, and α -particles at very low relative intensity (less than 1%). These radiations were found to decay with a half-life of about 4.5 hours and it was immediately assumed that the isotope was $^{244}\text{97}$ produced by the reaction: $^{241}\text{Am}(\alpha, n)^{244}\text{97}$. Soon

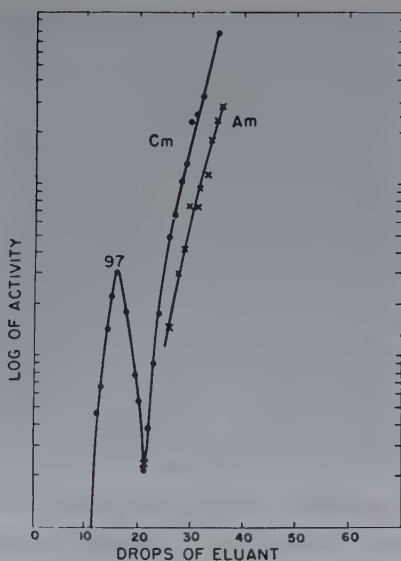


Fig. 5.3. Original elution data corresponding to the discovery of berkelium (^{243}Bk); S.G. Thompson, A. Ghiorso, and G.T. Seaborg, December 19, 1949; Dowex-50 eluted with citrate at 87°C .

thereafter it was correctly surmised that the main isotope, that giving rise to the observed α -particles, was actually $^{243}_{97}\text{Bk}$ produced by the reaction $^{241}_{95}\text{Am}(\alpha, 2n)^{243}_{97}\text{Bk}$.

It is interesting to note that experiments as early as the first day, i.e., Monday night, indicated that element 97 has two oxidation states, III and IV. The actinide concept provided the guidance to look for these two oxidation states, by analogy with the homologous element, terbium. In fact, the chemical identification procedure had been devised to accommodate either oxidation state and the large gap in the elution positions of element 97 and the curium was at first erroneously thought to be due to the fact that element 97 was in the IV oxidation state at that stage.

A name was given to element 97 immediately; in fact, you might say that a name had been chosen even before it was discovered.

According to the actinide concept, element 97 is the chemical homolog of terbium, which was named after the Swedish town of Ytterby. So the name "berkelium" after the city of Berkeley practically leaped out at us. I remember calling the mayor of Berkeley with the glad tidings and being very disappointed at his complete lack of interest. Stan and Al wanted to give berkelium the chemical symbol Bm, because it had been such a stinker in resisting identification for so long, but cooler heads prevailed and the symbol Bk was finally suggested and accepted by the scientific world.

Our suggested name didn't go uncontested. Two Soviet scientists, A.P. Znoyko and V.I. Semishin, in an article entitled "The Problem of Elements 97 and 98," published in *Doklady Akademii Nauk, USSR*,^{5,4} claimed the right to name element 97 on the basis that they had predicted its radioactive decay properties two years before its discovery using their Mendeleev periodic system of atomic nuclei. They suggested that it therefore be given the name "mendelevium" (symbol Md). Perhaps we satisfied these investigators some five years later when we ascribed the name "mendelevium" to element 101, on the sounder basis of recognizing Mendeleev's contribution to the chemical periodic system.

We had some more official communications concerning the spelling and pronunciation of "berkelium," since we preferred to keep the second "e" in the spelling, both of these as a faithful reflection of the tie-in to the name of the city, Berkeley. The nomenclature committee of the National Research Council suggested the dropping of the second "e," thus spelling and pronouncing it "berklium." The Commission on Nomenclature of Inorganic Chemistry of the International Union of Pure and Applied Chemistry suggested keeping the second "e," but pronouncing it "ber-ke'lium." As we know, the spelling remained with the inclusion of the second "e," i.e., "berkelium," but the pronunciation varies, with "berk'lium" prominent in the United States (as we prefer) and "ber-ke'lium" used more often in Europe.

As in the case of neptunium-plutonium, and americium-curium, where, in each case, the two members of the pair were discovered in



Fig. 5.4. Kenneth Street, 1950.

rather quick succession, the second half of the 97–98 pair was not long in coming. The reason for this is not difficult to understand — the background of preparations for element 97 applied almost equally well to element 98. In fact, there were no false lunges at element 98. The predictions of both its radioactive and chemical properties were made with uncanny accuracy and led us to our prey without a single misstep. Ken Street (Fig. 5.4) joined in this effort and perhaps this is the reason why no unexpected difficulties were encountered.

The first synthesis and identification of element 98 took place on Thursday, February 9, 1950. A target containing 8 micrograms of ^{242}Cm was bombarded with helium ions in the 60-Inch Cyclotron. A simple adsorption–elution separation was made using an ion exchange resin and element 98 was found to elute ahead of berkelium and curium, in sequence, and in the position expected. Actually, on the basis of column calibration experiments, element 98 was expected to elute onto collection plate No. 13 in the 26th and 27th drops of eluent, and this is exactly where it was found after a total elapsed chemical separation time of two hours. And it was expected to decay by the emission of α -particles of energy about 7.1 MeV, and

this is exactly what Al saw with his preset pulse analyzer. The half-life was found to be 45 min, also consistent with predictions.

This spectacular success of prediction of both the chemical and nuclear properties made possible an extraordinary sensitivity, for that time, for detection of such a small amount of α -radiation (about 80 α -counts per minute at the time of detection) present originally with over 10^{10} α -counts per minute of curium-242. This was probably the earliest date that our techniques had advanced to the point of making this possible. In fact, the identification of element 98 was accomplished with a total of only some 5,000 atoms; someone remarked at the time that this number was substantially smaller than the number of students attending the University of California at Berkeley.

The elution curve, corresponding to a subsequent bombardment is shown in Fig. 5.5, in which the elution positions of yttrium and berkelium are also included for purposes of comparison. The isotope

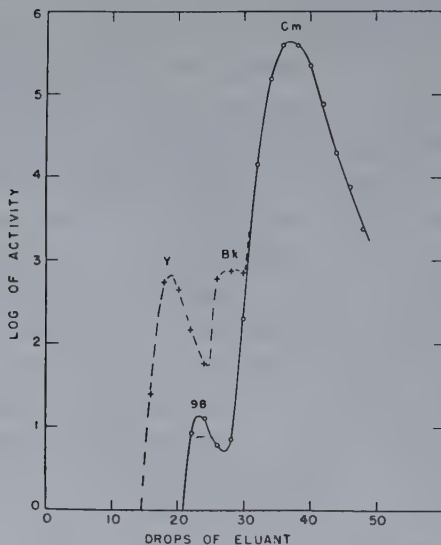


Fig. 5.5. Original elution data corresponding to the discovery of californium (^{245}Cf); S.G. Thompson, A. Ghiorso, K. Street, Jr., and G.T. Seaborg, February 9, 1950; Dowex-50 eluted with citrate at 87°C .

was thought to be $^{244}_{98}\text{Cm}$ produced in the reaction $^{242}_{98}\text{Cm}(\alpha, 2n)^{244}_{98}\text{Cm}$ but later work showed it to be $^{245}_{98}\text{Cm}$ from the reaction $^{242}_{98}\text{Cm}(\alpha, n)^{245}_{98}\text{Cm}$.

The naming of element 98 was not so straightforward as that of element 97. According to the actinide concept, element 98 is the chemical homolog of the lanthanide element, dysprosium. A little research turned up the information that dysprosium was named on the basis of the Greek word *dysprositos*, meaning "hard to get at." Although element 98 was hard to get at, in the sense that we had been working toward its synthesis and identification for some three years, we had trouble finding an analogous name.

Despairing of using the naming of dysprosium as an analogy, we toyed with a rather long list of possibilities. Upon looking through some of my old files I came upon a handwritten list of names that we apparently had under consideration for element 98: lewisium, cyclotronium, cyclonium, euprosium, nonactinium, enneactinium, enactinium, lawrencium, radlabium, praedicium, accretium, colonium, californium. As you know, californium (symbol Cf) won out, honoring the state and the university where the work was done. In a weak attempt to add justification by comparison with its homologous element, in our publication announcing the discovery of californium we added, "The best we can do is point out, in recognition of the fact dysprosium is named on the basis of a word meaning 'difficult to get at,' that the searchers for another element a century ago found it difficult to get to California."

Again, the naming of element 98 did not go unnoticed by those interested in this game. The newspaper writer Jack Wilson said in the *Des Moines Register*, for example, that it looked like the scientists had about used up the atomic possibilities of Berkeley, California. He said he would personally like to see what they would name an atom if they found one in, for instance, Vinegar Bend Alabama, or Pysht, Washington.

And the *New Yorker* magazine didn't remain silent. Upon learning about the naming of elements 97 and 98, the "Talk of the Town" section had this to say:

"New atoms are turning up with spectacular, if not downright alarming, frequency nowadays, and the University of California at Berkeley, whose scientists have discovered elements 97 and 98, has christened them berkelium and californium, respectively. While unarguably suited to their place of birth, these names strike us as indicating a surprising lack of public-relations foresight on the part of the university, located, as it is, in a state where publicity has flourished to a degree matched perhaps only by evangelism. California's busy scientists will undoubtedly come up with another atom or two one of these days, and the university might well have anticipated that. Now it has lost forever the chance of immortalizing itself in the atomic tables with some such sequence as universitium (97), ofium (98), californium (99), berkelium (100)."

The discoverers sent the following reply:

"'Talk of the Town' has missed the point in their comments on naming of the elements 97 and 98. We may have shown lack of confidence but no lack of foresight in naming these elements 'berkelium' and 'californium.' By using these names first, we have forestalled the appalling possibility that after naming 97 and 98 'universitium' and 'ofium,' some New Yorker might follow with the discovery of 99 and 100 and apply the names 'newium' and 'yorkium.'"

The answer from the *New Yorker* staff was brief:

"We are already at work in our office laboratories on 'newium' and 'yorkium.' So far we just have the names."

Before going on, I want to acknowledge the help of Burris Cunningham, Ken Hulet, and Gary Higgins in these experiments and in the preparation for them. Also critical to the success of the program were the contributions of Joe Hamilton, Bernie Rossi, Tom Putnam, and M.T. Webb in operating the 60-Inch Cyclotron in the Crocker Laboratory, and of Nelson Garden, Bill Ruehle, C.M. Gordon, and J.M. Davis for providing and supervising the use of the excellent

protective equipment for handling in a safe manner the high levels of radioactivity involved.

The discovery and early subsequent investigation of a synthetic chemical element, of course, involves work with tracer amounts. The second-most-important milestone in an element's life, after its discovery, is its first isolation in a ponderable, i.e., macroscopic (usually visible or weighable), quantity. We had to wait until 1958 for this to happen to both berkelium and californium. In that year Burris Cunningham and Stan Thompson succeeded in isolating for the first time compounds of berkelium, as the isotope berkelium-249, and californium, as a mixture of the isotopes californium-249, -250, -251, and -252. These had been synthesized in the intervening years by the long-term irradiation of plutonium-239 and its transmutation products with neutrons in the Materials Testing Reactor (MTR) at the National Reactor Testing Station in Idaho.

The first compound of californium of proven molecular structure (by means of X-ray diffraction) was isolated in 1960 by Burris Cunningham and Jim Wallmann, in the form of three-tenths of a microgram of californium (as californium-249) oxychloride. The pure

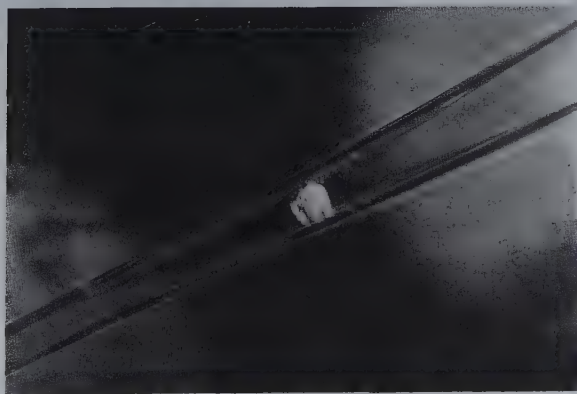


Fig. 5.6. The first pure californium compound isolated in 1960, magnified about 150 times (0.3 μg of californium as the oxychloride, CfOCl). The californium was in the form of the isotope ^{249}Cf . The crystals are lodged in a capillary tube.

oxide and trichloride were also prepared at that time. Figure 5.6 is a photograph of this first californium oxychloride.

The first compound of berkelium of proven molecular structure was isolated in 1962 by Burris and Jim. They isolated about 0.02 micrograms of berkelium (as berkelium-249) dioxide and used about one-fifth of this, about 0.004 micrograms (i.e., 4 nanograms), for the determination of its molecular structure by means of the X-ray diffraction technique.



Fig. 5.7. The four codiscoverers of berkelium and californium in Glenn Seaborg's office, Lawrence Berkeley Laboratory, January 20, 1975. L-R: Kenneth Street, Jr., Stanley G. Thompson, Glenn T. Seaborg, Albert Ghiorso.

Besides some further reminiscences by Stan, Al, and Ken (Fig. 5.7), today we will hear about much of the important research that has been performed on berkelium and californium during their lifetime of a quarter century. We will hear about their nuclear properties and chemical properties, about their production in astounding quantities, and about potential practical applications, both undreamed of 25 years ago.

5.2.2. Stanley G. Thompson

5.2.2.1. *Introduction: Glenn T. Seaborg*

So with that I'll go next to the first of the following reminiscers, Stan Thompson. I'd like to say a few words about Stan before I ask him to come up here. I don't know whether I should admit this or not, but Stan and I have been friends about 50 years — it'll be 50 years this fall since we met each other as freshmen at a high school in the Watts district of Los Angeles. Stan got his A.B. in Chemistry from UCLA in 1934 and came up to the Bay Area and worked at the Standard Oil Company in Richmond until 1942, when he joined me as a part of my Chemistry Section at the Metallurgical Laboratory, where he worked until just after the end of the war. By the way, he was there responsible for the conception of the process that was used for the isolation of plutonium in the Hanford production plant — the bismuth phosphate process. Here at Berkeley he's the codiscoverer of the elements with the atomic numbers 97 through 101 — berkelium, californium, einsteinium, fermium, and mendelevium. He participated, as I said, in the first isolation of microgram quantities of berkelium and californium. He has done extensive work on fission and most recently he has been carrying out research at the SuperHILAC on heavy ion reaction mechanisms. He was awarded the American Chemical Society's Award for Nuclear Applications in Chemistry in 1965. And with that, I'd like to ask Stan to come up to the lectern.

5.2.2.2. *Reminiscences: Stanley G. Thompson*

Well, I see a lot of old friends here today, people who were with us at the time of the early experiments we did when we first came to Berkeley. I see Herman Robinson, and Rosemary Barrett, and a number of other people. But, as Glenn said, a lot of work went into the discovery of these elements. I think the beginnings took place at the Metallurgical Laboratory in Chicago in 1945. As you may remember, the War was over in August 1945 and even by that time we

had started to do experiments in preparation for the attempt to produce berkelium. In fact, the first experiments were done near Christmas time 1945.

Some other things we managed to accomplish at the "Met" Lab in Chicago were to arrange to get samples of americium and plutonium in the Hanford reactor for neutron irradiation to make isotopes that were useful later in the experiments we did at Berkeley. Of course, we also had a lot of experience in separating actinides from other elements and fission products, etc., as a result of our work on the Hanford separations process. We actually did the first ion exchange separations at Chicago — although they were rather crude compared with the separations developed later. We also had some notions about how to handle radioactivity, which were put into effect later when we came to Berkeley.

Well, when we got to Berkeley the cupboard was bare, more or less. We had to start from scratch and build up a lot of equipment. And as Glenn said, we had a lot of help from people like Nels Garden, Red Gordon, John Gifford, Bill Ruehle, and others in designing and constructing equipment to handle radioactivity. We were successful in getting the gloved boxes, which are even used today; also what we called "junior" caves, in which we made the separations behind a moderate amount of shielding and handled the radioactive material with tongs. We designed and obtained a thick-walled lead cave in order to do the really high level separations. We worked over the top of this cave with tongs and attempted to see what we were doing with mirrors overhead.

Extensive work was also done to improve ion exchange separations. I think Ken Street and Gary Higgins did a tremendous amount of work on that, finally making it possible for us to do the separations of the individual actinides using cation columns operating at elevated temperatures. Of course Ken also had great success in developing concentrated HCl cation column separations of the lanthanides and the actinides which were absolutely essential in our work on berkelium. Ken Hulet gave us very valuable help with the separations, especially at the time of the discovery experiments.

In addition, a considerable amount of work was done in trying to predict the properties of the isotopes we attempted to discover. We worked on closed cycles and calculated masses, energies and half-lives. We used systematics, α half-life energy relationships for isotopes of the different elements, and even developed some rather crude electron-capture systematics. In fact, the measured half-lives of the isotopes we finally discovered were not too far from the predicted values.

So, little by little during that four-year period, we were able to develop the tools for doing the final successful experiments. I could tell a lot of funny stories — in a few years (like those when you work with people in a group such as ours with the people we had supporting us and helping with the experiments) a lot of amusing things are bound to happen. As one example, in the early days, about 1947, we attempted an experiment in which Burris Cunningham was working with us. Burris later on decided to drop out of the work on the new elements because of an extremely heavy load of other work. But, to return to the story, in those early days we attempted to take advantage of the expected +4 state of berkelium. We thought berkelium would have a +4 state about like cerium does, and we hoped to carry it in this state away from the americium target material using carriers like zirconium phosphate, bismuth phosphate, and ceric iodate. Well, actually, as it turned out, these separations were too slow and too inefficient. In this particular experiment Burris and I worked about 36 hours straight doing these cycles and trying to get something out we could identify, but without success. We were extremely tired — then we went to look outdoors and see what the weather was like. It was dark, it was cold, it was raining and windy, so I went to look for my coat and Burris helped me look. We searched for a long time and still we couldn't find my coat. Finally I happened to take a look at Burris and discovered that he was wearing my coat!

Well, all of these things together — and I would say it was a great team effort — resulted in the final experiments that were successful. When we did them, with the help of a good many excellent people,

it didn't take us more than a few minutes to be rather sure that we had climbed the mountain, so to speak. Those days in fact were very exciting and I, for one, wouldn't mind having them back again.

5.2.3. Albert Ghiorso

5.2.3.1. Introduction: Glenn T. Seaborg

Now I'd like to call on Albert Ghiorso after a few words of introduction. Al graduated from Berkeley with a B.S. in electrical engineering in 1937. During the following years, he more or less contributed his services working for a manufacturer of Geiger counters in the area, the firm that supplied us with some of our Geiger counters at that time. After I'd gone to the Metallurgical Laboratory in the spring of 1942 I got a letter from Albert asking whether I would be willing to recommend him for some kind of a job in the Navy — I don't remember just what it was — but I knew enough about Al to realize that probably wasn't the place for him. Actually, I didn't know Al very well. This is one of those cases where our wives took over. My wife, Helen, who had worked in the Laboratory here, as Ernest Lawrence's secretary, was a very good friend of Al's wife, Wilma, who was working here in the Laboratory as Donald Cooksey's secretary. When the letter came, Helen told me, "You hire this guy." So I wrote a letter back, enclosing a letter of recommendation for the Navy, but telling him that I would very much like him to come and work for us. He wasn't easy to convince; he was afraid that all I had in mind was for him to continue building Geiger counters. I assured him that that wasn't the case, that I had in mind a research position, although he still claims that the first six months or a year or so he spent a lot of time building Geiger counters. Anyway, he worked for the four years or so that we were at the Metallurgical Laboratory, participated, as you know, in the discovery of americium and curium (elements 95 and 96) there, and then returned with a group of us to the Radiation Laboratory at Berkeley in the spring of 1946, where he has been ever since. Al has been awarded an honorary Doctor of

Science degree from Gustavus Adolphus College in 1966; he's a recipient of the ACS Award for Nuclear Applications in Chemistry in 1973; and he is the codiscoverer of the transuranium elements with atomic numbers 95 through 106. I won't try to name them — I'd probably get mixed up. Al, would you come forward...

5.2.3.2. *Reminiscences: Albert Ghiorso*

The trouble with the 25th anniversary is one of perception — that our colleagues may grow old but we know that *we* don't really change! We look at one another and year by year things seem pretty much the same and then someone comes up, like Rosemary Barrett, and shows us a picture of what we looked like 25 years ago — there was a time warp of some kind, I'm afraid!

I don't have any particular prepared remarks. I wanted to listen to what Glenn and Stan had to say and perhaps fill in a few gaps. One of the things that are obvious looking at this early work is the relative crudity of the equipment that we used. This was simply because it hadn't been developed yet and we were just learning how to use it.

In the early experiments where Burris participated, Burris and Stan would work very, very hard with a tremendous number of separations, very difficult procedures, and end up with a small sample. They would hand it over to me and say, here, we're tired, you find out what's in it. And usually there would be a few α -counts of something. One time in particular, I remember, where we had 8–10 counts per minute of what looked like curium-242 from an α -pulse analysis. Our accuracy wasn't that great, so I followed decay on it day after day and sure enough at first we were very excited to find that the points indicated a half-life different from that of curium-242. So we kept on counting it for another few months and then... you know the answer, of course — it *was* curium-242.

After some months it became obvious that we had to have detection of Auger electrons from electron capture to get more sensitivity compared to counting X-rays. I worked on thin-window counters,

which were a bit unstable in those days, but I was never really sure that what was being counted was Auger electrons. I could always accuse Stan and Ken of not having a clean sample and we had a lot of false alarms; then when we really did find the electrons from 97-243/4 it seemed almost unreal.

We also used an Eck and Krebs commercial Geiger tube filled with xenon, to look at L-X-rays and we actually did show the difference between 96 and 95 X-rays — in other words, L-X-rays from the decay of 97 which were curium-characteristic X-rays. We actually showed that these were curium X-rays in our first paper, which was very nice.

We used a gridded ion chamber to observe α -particles. We knew by this time that α -particles were really the thing to go after, and they are still my favorites because an α -particle is so distinctive it's almost like giving an atom a name. There aren't too many overlaps and we are able, by being very careful, to get half-lives and energies to pinpoint the identifications of atoms. Our early work starting in 1946, when a number of us came back from Chicago, was pretty much devoted to exploiting what we could do with the 184-Inch Cyclotron. It was working very well, and we (Wayne Meinke and I) would bombard targets of thorium and uranium and make a lot of spallation reactions. The result was that we came up with a whole new series of isotopes, the collateral radioactive families, and got to be pretty good at it. Even though our methods were crude, we were able to show the difference from one series to another; it amounted to working on perhaps 30 to 40 different α -emitting nuclides. We became fairly familiar with a technique that got to be pretty powerful.

The name of the game, however, was to do it at a very low level with very high sensitivity against no background. Well, in those days things were somewhat crude, so you couldn't do the nice experiments that one can do now where you can look at literally one α -particle that decays perhaps in months and be sure that you're not making a mistake. At that time we weren't really confident all the time, but we worked at it and of course the techniques became very

valuable. I remember the particular day when we saw the three α -peaks which characterize the α -particle fine structure of berkelium-243 — they stood out loud and clear. For a while we thought we had two different isotopes. But these three peaks which we saw — an intensity of some counts per minute — you could just take a look at them and glory that these counts were there.

The instrument that we used was a 48-channel instrument — 48 whole channels of pulse analysis. That was the biggest in the world but it was kind of crude. It was developed originally in Chicago by Herman Robinson and some other people and we had reproduced an advanced copy here at Berkeley. So here we had these 48 channels; each one of the channels drove a mechanical register directly. There were no scalers, so the counting rate was limited, but the counting rates we had were never very high, so it was not any great problem. Here we had these dial-like instruments, arranged in rows on a rack one after the other, and at first we used to reset them to zero by hand. It was quite a job to use your fingers and set them all back to zero — you got to be pretty skillful. But we got pretty tired of that and Herman thought, well, let's have a little bit of advanced technology, and so he had the registers changed slightly and devised circuitry to set them back electrically. His big power supply just drove them at 60 Hz till each came to its mechanical stop. After we were through with recording our spectra we would press a button and the registers would all go whizzing back — it was quite a spectacle — until they stopped at zero. The power supply was on top of the rack that held the registers, a great big thyatron rectifier power supply right in the open, about 440 V of pulsating direct current going into these 48 registers. It was a lot of power.

Well, that worked well, but a few years later it got even with me, I'm afraid. Something went wrong with the system and I was standing on a stool in front of the rack trying to troubleshoot the problem. Somehow I was holding onto the rack with one hand and my other hand touched a part of the power supply — I violated my rule, and everyone else's rule, never to use more than one hand

in any place where you might have a problem — and I was frozen in place, and the hell of it was that I knew I was frozen — I couldn't get off. I could literally see myself being electrocuted. It was frightening! Fortunately Herman had thought ahead. Right up there at the top of the rack was an "off" button and an "on" button. I slumped on the stool and my breast hit the "off" button and I fell to the floor. History would have been a little different if that button hadn't been there, I assure you, because I was well on my way to being electrocuted. I'll never, never forget that for the rest of my life. This happened a few years later than the discovery of 97 and 98, just about the time we were doing the experiment on element 101.

[Hyde calls out, "A round of applause for Herman!" and the audience responds.]

The work was difficult and it was primarily a chemical accomplishment. The counting was sort of anticlimactic — the real work had to go into the chemical separations necessary to isolate the new atoms from these difficult bombardments. This was the time when chemistry really paid off, and I think Stan and Ken deserve the plaudits for their tremendous accomplishment, because it really was a chemical achievement at that time. As we proceeded to higher elements it became more and more a physical thing, until today we can't do chemistry fast enough. That is something that we hope to do in the later stages of development of the experimentation.

Well, this reminds me of the fact that this is another anniversary. Twenty-five years ago we started doing heavy ion work at the 60-Inch Cyclotron. There weren't any heavy ions up to that time. I remember going down to the campus once a week, working with Bernie Rossi, to develop ion sources that would give us usable heavy ion beams. We made bombardments and discovered how to make exotic ions as heavy as oxygen — as high as we ever got. That led to plans eventually where Luis Alvarez proposed a HILAC which led to the Omnitron and then to the SuperHILAC. So to me it's been a relatively short period. It doesn't seem like 25 years and yet obviously it has been; it's been a very rewarding period for me.

5.2.4. **Kenneth Street, Jr.**

5.2.4.1. *Introduction: Glenn T. Seaborg*

Ken Street holds the unusual distinction of being a native of Berkeley, California. He was born in Berkeley and he got his B.S. in Chemistry here at the University in 1943, then he went into the Marine Corps, came back to the University and did his research, his Ph.D. work, here in the Radiation Laboratory. Actually, he was one of my students. I'm very proud to say that he got his Ph.D. under me. That doesn't mean I taught him anything. When you have a man like Ken Street, he teaches you more than you ever teach him. He joined the faculty; of course, we recognized that we had a good man and we immediately tried to persuade him — and succeeded — to become a member of the faculty at Berkeley in the Chemistry Department. And he was a member of the faculty until about 1952 or so, when he began an association with the Livermore Laboratory, now the Lawrence Livermore Laboratory; he served in various capacities there as Chemistry Division director, and so forth, on up the line, until 1958–59 when he was Deputy Director of the Livermore Laboratory. Then he returned to Berkeley as a full Professor of Chemistry in 1959, and at all times while he was at Berkeley he did his work in the Radiation Laboratory, intimately connected with the Berkeley campus. In 1974 Ken went back to the Lawrence Livermore Laboratory as Associate Director for Energy and Resource Programs. His research interests have been in nuclear chemistry, molecular spectroscopy, and geochemistry. Ken...

5.2.4.2. *Reminiscences: Kenneth Street, Jr.*

I must say, a few weeks ago when Glenn called me and told me he was planning a little session on reminiscences to celebrate the discovery of berkelium and californium, I had more in mind a few of us gathering around a fire with a beer or two, telling a few dirty stories and recounting some of the minor felonies we committed in the course of getting the job done. When I'm confronted with

microphones and cameras — my instinct of preservation says I should give my name, rank, and serial number and sit down. Actually, I regard myself as being very fortunate to be here and to have been a part of this work. I was not a member of the Chicago Mafia that came west after the war and had been working on these things and continued for a long time afterwards. I am sort of a transient in the transuranium business — I was in it for only a period of about four years and have been doing other things since.

Glenn asked me to say a little bit about the development of some of the ion exchange chemistry that went into the final solution of the problem of getting berkelium and californium out in a form in which we could identify them. I started in this business as a graduate student when I came to work for Glenn in 1946. As with most of us in those days, I owe a great debt to Burris Cunningham, too, for pointing me in this direction to solve a number of the problems that we had at that time. We also had a lot of help later on, as Stan and Glenn have mentioned. Gary Higgins and I, Ken Hulet and Stan, all worked very closely together in developing these techniques.

My first problem when I arrived up the Hill with Glenn was to find some way of getting a reasonable separation of americium and curium from the rare earth fission products. I was following in the footsteps of some people that some of you know, most of you probably — Ralph James and Tommy Morgan, who had been working on americium and curium isotopes with Glenn both in Chicago and back out here. They had identified most of the longer-lived α -emitters and my task was to carry on this work with the lighter isotopes of americium and curium. These were all expected to decay by K-electron capture to a large extent, and we might very well be much more interested in the electromagnetic radiation than had previously been the case. So I set about to see if we could find some way to separate the actinides from the rare earths and I must admit that my start on this was mainly a matter of desperation. I had to do it one way or another and we would have settled for almost anything. The early work was directed at simply finding a way, we hoped, to move the actinides enough with respect to the rare earths

so that the actinides elements like americium which overlapped promethium on the column which was eluted with citrate ion — which was the common technique then — would be moved at least a little bit so that by doing two ion exchange runs we could separate the actinides from the rare earths.

I'd like to be able to say we had a very carefully laid out plan based on fundamental theory and using the latest techniques and extrapolations of well-known and established principles to do this work. As a matter of fact, I think our procedure much more closely paralleled a dictum that Earl Hyde has often quoted to me, "If you root around under an oak tree long enough, you stand a pretty good chance of coming up with an acorn." A number of us started rooting pretty hard and we didn't go very far looking for things to try. We used almost anything that was at hand, starting with very exotic reagents like nitric, phosphoric, sulfuric, and hydrochloric acids. Fortunately things turned out much better than we had any reason to expect. We very early discovered that not only did HCl, even at concentrations as low as 6 M, displace the actinides with respect to the rare earths sufficiently far that you could clearly make a separation in two ion exchange runs, using both citrate and HCl as eluants, but we also fell into very good fortune and when we kept pushing the concentration of HCl higher and higher — and, as I recall, we got all the way up to ~13.5 M and then started pouring in alcohol to increase the activity still further — the whole actinide group moved up and eluted well ahead of lutetium so we had a very rapid group separation of the actinides from the rare earths.

I'd like to say that there were a number of people who contributed to this and also to the work that was done later in order to calibrate the citrate columns so that we could make the final runs in December that resulted in the identification of californium. I think, like my colleagues have alluded to previously, all these things come after a lot of people have made a lot of contributions that finally stack up enough of the blocks so that you can put the whole thing together. I regard myself as being extremely fortunate to have been around and, as Glenn said, maybe I was waiting for this — I was

born in Berkeley and have been here most of my life. But I was here at the right time and the right place and so got to take part in this very exciting adventure.

Seaborg: Ken, you didn't make it very clear where you poured that alcohol."

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Chapter 6

The “Big Bang”: Discovery of Einsteinium and Fermium

Although most of the discoveries of the transuranium elements were carefully planned based on the best available knowledge of chemical and nuclear properties, element 99 (einsteinium) and element 100 (fermium) were synthesized in a most dramatic, unplanned, and unexpected manner in the first U.S. thermonuclear device, “Mike.” It was designed and tested by the Los Alamos Scientific Laboratory (LASL) at Elugelab Island in the Eniwetok Atoll of the Marshall Islands in the South Pacific on November 1, 1952. Because of the enormous nearly instantaneous high neutron flux ($\approx 10^{24-25}$ neutrons/cm²!) generated by the ≈ 10 -megaton detonation, at least 17 neutrons were successively captured in the ^{238}U in the device to produce isotopes through ^{255}U with enough yield to detect the more stable higher Z elements produced by subsequent β -decays (Fig. 6.1). The discoveries occurred only because chemical and isotopic identifications were made routinely at LASL in connection with these tests in order to establish the properties, efficiencies, and performances of the explosion devices. Accordingly, debris for this purpose was collected on filters which were flown through the explosion area. Naturally enough, the first recognition that something extremely unusual and unexpected had happened came from LASL.

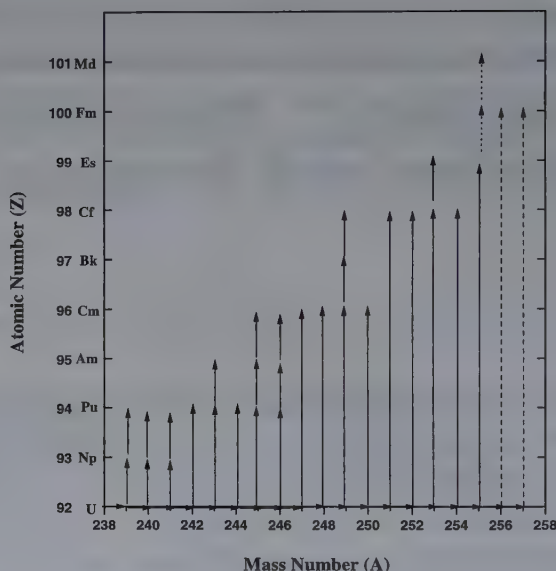


Fig. 6.1. Schematic of production of heavy uranium isotopes by successive neutron captures in ^{238}U , followed by their subsequent β -decay.

6.1. The View from Los Alamos

At the time of the test, Dr. Roderick ("Rod") W. Spence was the Group Leader of the LASL Radiochemistry Group J-11, which was responsible for the radiochemical analysis of the debris from the test in order to determine details about its yield and performance. Rod was staying on the Kwajalein Atoll, 300 miles from ground zero, during this period of time. At the 25th Anniversary Symposium commemorating the discovery of einsteinium and fermium, he recalled^{6.1} details of how the Mike device looked (Fig. 6.2) on a tower prior to the explosion and showed a "before" picture of Elugelab and the Mike device (Fig. 6.3). Figure 6.4 shows an "after" picture in which Elugelab as well as the Mike device is completely gone! An early photo of the actual Mike explosion taken from some miles away and another photo taken somewhat later from the photographic plane which Spence was on are shown in Fig. 6.5. Although

the photographic plane was supposed to get no closer than about 300 miles, he thinks they "fudged" a bit and were probably only 75 miles away. Estimates of the magnitude of the explosion can be made from the size of the stem and cloud. At much later times the cloud was some 300 miles in diameter and the stem some 30 miles in diameter!



Fig. 6.2. Ivy-Mike Device on Elugelab. Note its size compared to the vehicles.



Fig. 6.3. Photo before the Mike explosion. The Mike device and Elugelab can be seen at the top center.

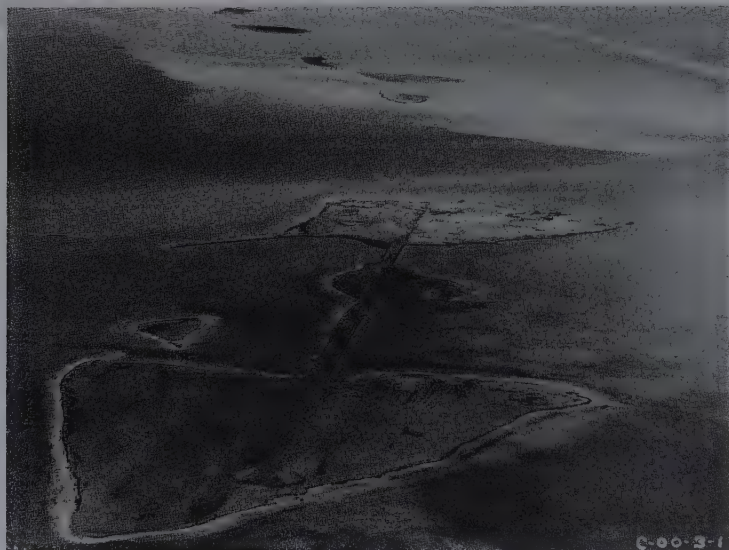


Fig. 6.4. Photo after the Mike explosion. Elugelab is gone!

(a)



Fig. 6.5. (a) Early photo of Mike explosion taken from a few miles away. (b) Later photo of the Mike "cloud" taken from the photographic plane that Rod Spence was on, only about 75 miles away.

(b)



Fig. 6.5 (*continued*)

In order to get more precise measurements of Mike's yield and performance, samples of the cloud were collected using F86 fighter planes. These planes flew through the cloud carrying special filters attached to their wing tanks, a technique which had been worked out to collect samples from previous atmospheric tests in the Pacific in 1951. For more details on the logistic difficulties of refueling the planes, etc., see Spence's more detailed account in Ref. 6.1.

The precious filter papers were divided into two shipments and flown in separate planes to Kirtland Air Force Base in Albuquerque, New Mexico, to ensure that at least one set would arrive safely. At Kirtland they were transferred to smaller planes to be flown to the 7200-foot-high airport at Los Alamos, New Mexico. The filter papers were taken to the old J-2 building, close to where the Los Alamos Inn is now. The filters were typically dissolved in fuming nitric and perchloric acids to give clear solutions, which were then analyzed for uranium, neptunium, plutonium, and fission products. However, these filters were so heavily contaminated with coral (CaCO_3) from the Atoll that serious problems were encountered in dissolving

them; many samples literally caught fire, presumably because of the chemical reactions taking place due to the large amounts of coral. At Los Alamos, Ed Moore, the analyst for plutonium in Spence's group, was having much difficulty decontaminating it from the β -activity, which he presumed was due to fission products although he had never had this trouble before. Charles I. Browne and H. Louise Smith then took up the problem of trying to isolate and α -count the plutonium fraction from the samples, but encountered large amounts of inexplicable β -activity, which turned out to be from a new plutonium isotope, ^{246}Pu . (Charles had only just come to Los Alamos as a military staff member in September 1952 — he had received his Ph.D. in Nuclear Chemistry at UCB in 1952 in Isadore Perlman's group. And Louise had only been at the Laboratory a week, her first job after graduate school.)

A picture of Browne, Smith, and Spence on the occasion of the 25th anniversary of the discovery is shown in Fig. 6.6. In the words of Louise Smith, "We thought our chemistry was wrong, but it wasn't. We were seeing new plutonium isotopes which were characterized by β - rather than α -activity." Browne recalls taking the purified plutonium fraction and "milking" (separating) the americium daughters which were being formed by the new β -decaying plutonium isotopes. The samples had to be taken to another site in the Laboratory for spectral analysis of the radiations using equipment (an old 10-channel pulse analyzer) which only became available during the evening hours when it was not needed in the production effort. Smith and Browne subsequently identified the new β -emitting isotope as Pu-246. Smith also recalls the "mysterious alphas" with energies of 6.6 MeV and 7.1 MeV, which they were only able to say belonged to elements with Z greater than plutonium, i.e., transplutonium elements. They had done very little research on the heavy elements and had no equipment for that purpose, although they did typically separate transplutonium fractions. To Darleane Hoffman's eternal regret she was unable to participate in these efforts due to her "misplaced" clearance which delayed her from joining the Los Alamos group from January 1953 to mid-March 1953. But one of her



Fig. 6.6. Charles Browne, Louise Smith, and Rod Spence, the three LASL scientists involved in the discovery of einsteinium and fermium, reminiscing on the 25th anniversary of the discoveries.

first memories after joining the Los Alamos group was of seeing these large coral samples being dissolved outside, away from the building, in a makeshift tent laboratory!

Although the primary job of the LASL group was to determine the yield and efficiency of the Mike device, it was their early discovery of at least one new β -emitting isotope of plutonium during the analysis of the debris that told them that something very unusual and unexpected had occurred. This led to the subsequent discovery of the new, very neutron-rich isotope ^{244}Pu in the mass-spectrometric analyses by ANL scientists along with their corroboration of the LASL discovery of ^{246}Pu . ^{244}Pu is a very important plutonium isotope since it is now known to have a half-life of about 80 million years, barely long enough for it to be found in nature. (Some 20 years later, as described in Chapter 2, Darleane Hoffman and coworkers proved that this was the case in a very difficult and elegant experiment.) The Argonne and LASL teams later identified a third new plutonium isotope, ^{245}Pu .

A couple of months later, after the discovery of element 99 at Berkeley, as described below, it became clear that more material would be very valuable, so, at Seaborg's suggestion, Spence arranged to recover some tons of the coral. These were independently and laboriously processed at all three labs to gather more transplutonium resource material so that the bonanza of new research possibilities could continue.

6.2. The View from Berkeley

Ghiorso, drawing heavily on Seaborg's *Journal*,^{6,2,6,3} describes the sequence of events that happened with amazing speed at Berkeley when the famous teletype message was delivered announcing the discovery of ²⁴⁴Pu in that explosion. It should be remembered that this was more than a month after the thermonuclear test. The rapid pace of the work at Berkeley is illustrated by the following dates taken from the *Journal*.

"December 4, 1952. The complete story of this famous event in the history of the discovery of the elements has never been told before. Our first hint that something very unusual had happened in the South Pacific came when we read the secret teletype message sent to Seaborg and Iz Perlman on December 4, 1952, from the office of James Beckerley, the Director of the Office of Classification [Ref. 6.2, p. 295], which is reproduced below:

'We are trying to withhold publication of further information on U-240 until we have had an opportunity to review entire field of isotopes important to LRD work. It is our understanding that Perlman is preparing for publication revised UCRL-1928 with some additional data on the isotope. Request that any new data be withheld from publication until further notice even though guide permits declassification. We expect to review heavy isotopes situation at January senior reviewer meeting and issue bulletin clarifying position.

Radiochemical data on the recent Eniwetok test indicates presence of some unique heavy element isotopes such as Pu-244.

We do not want to release any information on the properties of these isotopes, even though information is declassifiable under guide. Accordingly you are requested to withhold publication of any information on the existence and properties of isotopes present in debris samples and consider such information as secret, restricted data. This prohibition applies even when information is dissociated from test. Please inform those in laboratory who might have access to these data.'

"This message was quite intriguing since we had heard rumors about an H-bomb test. We found out later that its code name was Mike and that the huge explosion had taken place on November 1, 1952, in the South Pacific. Otherwise we knew nothing about it since everything was highly classified. We had no connection at all with the weapons test program, so there was no way for us to find out what had happened. The diagnostic tests for all weapons tests were handled by "Yellow Nose" groups at the Los Alamos Scientific Laboratory and the Argonne National Laboratory and not even their existence was made public.

"For a few years we had been pursuing a long range program aimed at producing heavier nuclides by the bombardment of plutonium with neutrons at the Materials Testing Reactor (MTR) in Idaho. This program had the descriptive name of "napkin ring" and had as its first objective the production of ^{244}Pu , the new isotope mentioned in the teletype. The Argonne Laboratory was well aware of this because it was using the MTR also to produce the same heavy nuclides; that is probably why Beckerley's communication was sent to us, just in case we had found the isotope in this way. And indeed this did happen a couple of years later. Now we were being told that this isotope had been produced in an instant! We found this information to be quite amazing but what followed was even more fantastic.

"I discussed the teletype with Stanley Thompson and found that he, too, was consumed by curiosity. By now, we told ourselves, surely the Los Alamos and Argonne diagnostic groups must have discovered more than ^{244}Pu . After all, it had been five weeks since

the explosion had taken place, plenty of time for them to have obtained much more information. However, since their work was entirely classified we knew that we would not be told very soon under normal circumstances and thus it would be a long time before we would find out what had been discovered. It was even surprising that they would tell us of ^{244}Pu . Maybe they had even found a new element or two! We felt that somehow we should join with them and help with the research or we would be left far behind in the field that we had started. But how could we do that? That night, at home, I made a very simple estimate, the wildest idea of my entire career, which, by sheer luck, turned out to be remarkably accurate.

"Here is how I made my estimate. I knew that ^{244}Pu could only have been detected with a mass spectrograph, so I assumed that the amount present in the plutonium fraction of the device was about 0.1% of the amount of ^{239}Pu (that was roughly the limit reached by mass spectrometers of the time — and by a strange coincidence that is what its abundance turned out to be!). I assumed that ^{238}U was present in the thermonuclear device as a major component and that somehow in an instant it had been subjected to an enormous neutron flux from the fantastic multimegaton explosion and would successively capture neutrons to momentarily create ultraheavy isotopes of uranium. Since these atoms would have a large excess of neutrons, they would be highly β -unstable and would quickly decay by β -emission to elements with more protons. I assumed that the yield decrease with mass would be logarithmic and that the yield line would be represented by my assumed $^{244}\text{Pu}/^{239}\text{Pu}$ ratio of 10^{-3} . The yield would thus decrease by a factor of a thousand for every increase in mass of five units. Finally, I assumed that about 10^{14} atoms would be available from the fraction of the bomb recovered; this guess was based on my experience gained two or three years before, when I analyzed some Soviet and American bomb debris. Putting these numbers together I speculated that 16 neutrons might be captured with a yield 10^{-9} of that of ^{239}U and that the ^{254}U thus formed would β -decay all the way up to atomic number 100! A reasonable prediction based on the systematics of that time was that

this nuclide would be an α -emitter with a half-life of about a month and even though its production had taken place more than a month earlier we might still observe something like an α c/m. This would be quite a step upward from the heaviest nuclide known at that time, ^{246}Cm !

"I found this extremely exciting and at the Lab the next morning I sought out Stan once more to see what he thought of this fantasy. He was immediately enthusiastic and suggested that he could probably get part of the filter paper that had been given to the fledgling Livermore Laboratory. The Livermore Laboratory had just been started by Ernest Lawrence and Edward Teller as a bomb laboratory in competition with Los Alamos and Ken Street had been appointed head of its Nuclear Chemistry Department. Ken had been our colleague in the discovery of element 98 just a couple of years before, so Stan had no trouble in enlisting his cooperation. In order for Street's group to practice their bomb analysis techniques they had received one of the filter papers that had been flown through Mike's mushroom cloud and Ken was willing to let us have half of it for our exploration.

"Stan felt that there would probably be problems in dissolving the filter paper but that this could be handled. Stan was a very innovative, bright chemist and had lots of experience in solving difficult chemical problems. At the Metallurgical Laboratory in Chicago, working for Seaborg in the early 40s, he had invented the bismuth phosphate process that was used so successfully at Hanford to separate plutonium from the fission products on a large scale and he knew that he could overcome any difficulties that they might encounter. He called it "fun" when he came up against a difficult problem. Seaborg had assigned one of his graduate students, Gary Higgins, to work with Stan and together they had worked hard to develop the use of ammonium citrate as an eluting agent for ion exchange columns to separate the chemical elements beyond plutonium. Now, with the valuable experience that they had gained, they felt that the necessary chemistry was well in hand. This was a particularly tricky bit of chemical wizardry but, after the discovery of

berkelium and californium a couple of years before, they had continued to work on their techniques and had improved them even more. They had complete confidence in Seaborg's prediction that elements 99 and 100 would have stable 3+ oxidation states and thus would adhere to their predicted elution positions.

"Stan and I felt that it was worth pursuing this very romantic idea, but what would the others say? When we consulted with Seaborg and his deputy, Iz Perlman, to see what they thought of the wild idea we were surprised to find that both of them were very skeptical but they did not try to dissuade us. Perhaps we should not have been surprised at their skepticism, since we were postulating that as many as sixteen neutrons would be captured in an instant during the explosion to convert ^{238}U into ^{254}U . They both felt that it would be impossible for a nucleus to bind that many neutrons and that, although they did not oppose it, they considered that any effort would be futile. (Future tests were to show that as many as nineteen neutrons could actually be captured to make the heaviest fermium of all that could be made in bomb reactions, ^{257}Fm !) The foregoing is not meant to denigrate Seaborg and Perlman; it is meant merely to show that the multineutron capture concept was not one that was intuitively accepted by everyone, even by the best minds of the division. Even the physicists who designed the bomb did not think of this possibility! They were just as surprised as everyone else.

"December 5, 1952. In the next morning's mail Glenn received a copy of a letter from Spof English to Doyle L. Northrup about the isotopes from the Mike debris which he showed to us. English included a copy of the telegram from Ralph Carlisle Smith (Los Alamos) to James G. Beckerley (AEC, Washington), dated December 2:

'At the request of Los Alamos, a sample of Mike shot has been analyzed and found to contain plutonium two four four. Other heavy element unique isotopes are also present in the samples. This information indicates an especially high neutron flux, which reveals something of nature of device detonated. Samples of the bomb debris have been made available to Argonne, Livermore, and AFOAT-I organizations. Therefore Bradbury, Kellogg, Graves,

and I recommend that you advise all organizations including Joint Task Forces that all analyses of the debris be considered part of the Ivy Report Series and graded Secret Restricted Data. Furthermore advise all concerned that the existence of plutonium two four four is considered Secret Restricted Data at this time in order to avoid an injudicious announcement by organizations which might acquire samples. If the source of the sample were not unique, it is understood that this type of information is declassifiable. Past experience shows that some organizations fail to distinguish between declassifiable and open fields, hence our request for the advisory communication.'

"This communication did not add anything new but it reinforced our feeling that we should do something to get involved. Fortunately, Stan and I were young — he was 40 and I was 37 — and we were not about to be deterred by what seemed like the impossible, since we felt that there was nothing to lose. We were pretty cocky and brash. After all, we had discovered elements 97 and 98 in exceedingly difficult experiments just a couple of years before, hadn't we? Maybe we would be lucky! With half of a filter paper and only a small effort we felt that we would at least find out what had been discovered by the weapons teams.

"With the helpful advice of Bill Crane, who had just transferred from Stan's group to Livermore, Stan and Gary went to work (around the clock) to separate out the elements beyond atomic number 94. As expected, the filter paper was very difficult to digest and took a lot of time. Incredibly, though, only two days after they began this dissolution I had begun to analyze their samples eluted with citrate from the cation exchange column using the same 48-channel α pulse height analyzer to analyze the pulses from the Frisch grid chamber that we had used for the berkelium and californium experiments just a couple of years before. A 2% aliquot of the transplutonium fraction had indicated a little earlier that there were interesting americium, curium, and californium alphas, so we went ahead knowing that the experiment was going to be worthwhile, but we were not prepared for the surprise that we now encountered.

"December 13, 1952. To our utter astonishment, within minutes it was obvious that there was about 1 c/m of an α -particle that we had never seen before! It had an energy of about 6.6 MeV and it eluted just ahead of a 6.1-MeV α -emitter that we assumed was coming from a new heavy isotope of californium. There were even a few spontaneous fissions and this seemed entirely reasonable for an even mass isotope of element 100; in fact, we used this as an argument that it was element 100 rather than 99. The fact that the new α -particle activity eluted just ahead of the assumed californium peak meant that it could be coming from either element 99 or 100. This particular ion exchange column had not run properly, so we could not be sure which it was. Gary kept insisting that the 6.6-MeV elution peak was too close to californium to be due to 100 and thus must be due to 99. But Stan and I kept pointing to the fissions as evidence for its assignment to element 100. The next citrate elution column separation would tell. The important thing was that the speculation had paid off and now we were really involved. Needless to say, Glenn was just as excited as Stan and Gary and I were and now became intimately involved in the details of the subsequent analyses. Glenn's office was just across the hall in Bldg. 5, where I and the grid chamber were located, so it was easy for him to watch over the latest developments. Of course, Glenn would keep careful track of the subsequent events in his *Journal* and these records would turn out to be historically valuable.

"After following the decay of the 6.6-MeV α -particles for a few days, we found that their half-life was of the order of a month, so now every aspect of the prediction seemed to have been correct. It was a spectacular proof of the whimsical, tongue-in-cheek "Ghiorso uncertainty principle" which I had been fond of quoting for the last ten years! In its simplest form it goes like this: *If one has enough uncertainties in the first measurements of something important, the correct answer will be obtained.* This was a classic case, indeed, because the only fact that I knew for sure in making my calculation was that ^{244}Pu had been formed in the first H-bomb explosion. Now it seemed very likely that we had discovered $^{254}100$. The rapid process that

goes on in the stars (the "Big Bang") had been reproduced on earth! We found this so startling that we decided that we had to prove without question that we had found a Transcalifornium activity by making a new run, this time with a 1.5-day ^{246}Cf tracer. We would generate this tracer by bombarding ^{238}U with ^{12}C ions at the Berkeley 60-Inch Cyclotron at Berkeley.

"December 15, 1952. Fortunately, Ken Street was able to obtain new material from Lloyd Zumwalt of Tracerlab, a small commercial company in Berkeley doing contract analysis of the weapons tests for AFOAT, the military agency that handled these matters. This time the procedure to make samples for pulse analysis was much faster because we got a lanthanum fluoride precipitate fraction containing the heavy elements rather than a filter paper. It was relatively straightforward, after a little chemistry, to load it and the ^{246}Cf tracer, onto a cation exchange column to separate the actinide elements.

"December 17, 1952. The pulse analysis began and again we were observing about an α c/m of the 6.6-MeV alphas. And once again they were eluting just ahead of the 6.1-MeV peak that had been produced by Mike, but this time we also had a ^{246}Cf tracer peak to confirm that the observed 6.1-MeV peak was due to californium. Now there was no doubt. We were looking at a new element and it had to be higher Z than $\text{Cf}=98$, presumably either 99 or 100!

"The question immediately arose: What had been found at the other two labs? Maybe we had just made an independent discovery. After all they had been working on the problem for more than a month with far more material (we found out later that they had more than an order of magnitude more!), so that they should have been able to find out what we had and more. On the other hand, neither of the other two labs had much experience in the very heavy element region and it was even quite possible that because of the press of their other work they had not even had time to look.

"December 18, 1952. Glenn, always with an eye for history, decided that we should go on record and suggested that Stan and I write a memo to him and Street on what we had found. That we did

and soon it was finished [Ref. 6.2, p. 310]. In it we demonstrated that we had observed a new transcalifornium isotope with an α -energy of 6.6 MeV, but the data were insufficient to distinguish between elements 99 and 100. We suggested a tentative assignment of the nuclide to $^{254}100$ because we had also seen a small amount of SF activity. (We did not realize until after the next experiment that the fissions came from the tail of the strong californium-254 SF activity.) Copies of the memo were immediately sent out to Los Alamos, Argonne, Armed Forces Office of Atomic Energy (AFOAT), and Livermore. In addition, Glenn made telephone calls to inform these people immediately of what we had found.

"His call to Rod Spence, the Radiochemistry Group Leader at Los Alamos, produced the information that his group had not yet investigated the heavy element region at all and he was quite surprised and congratulatory. When Glenn asked if we could get more material so that we could look for element 99, which at that moment we thought could be long-lived, he said that he had sent the "lion's share," essentially 90% of what they had, to Argonne so that the plutonium and curium fractions could be mass-analyzed. A call then to Winston Manning, the chemistry division director at Argonne, also indicated surprise at our findings. He congratulated us and said that he would take Glenn's request for more material "under consideration."

"December 20, 1952. Since we knew that it might take a little time to get material from Argonne, we looked around for other sources. In the meantime we redissolved the eluted material from run No. 2 and eluted it from another cation exchange column. On December 20 the column was run, this time under exactly the same conditions that we had used in separating element 98 in the discovery experiments of 1950, so that we could better define the atomic number of the 6.6-MeV α -activity. We found that Gary was right; the activity was due to element 99 rather than 100 and the fissions were coming from californium. In addition, we found the 6.6-MeV α -activity was growing into the newly purified californium fraction as well as decaying in the element 99 fraction, and that fact made its identifi-

cation absolutely certain. Its californium β -emitting parent turned out to have a half-life in the month range also. Unfortunately, we did not communicate this new information by telephone immediately and that was a mistake.

In his *Journal* entry [Ref. 6.2, p. 326] for December 20, 1952, Glenn wrote:

'We decided not to bother writing a correction to yesterday's memorandum (because of the admonition for the maintenance of secrecy), but to tell the interested parties when we see them or have phone conversations with them (and this proved later to be an unfortunate decision).'

"January 7, 1953. In a couple of weeks we got an answer from Argonne. But it was not in the least what we had expected! Instead of additional material we got a memo given to Seaborg when he happened to pass through Chicago. He was handed a report, ANL-WMM-1059, dated the previous day, January 6, 1953. It suggested that the assignment of our 6.6-MeV alphas should be to an isotope of element 99, not 100; in addition, the Argonne group claimed the discovery of a "6.35 MeV" α -particle activity, which they assigned to element 100! This activity, which Berkeley had not seen, had an abundance of about 1/5 that of the 6.6-MeV alphas and did not seem to be decaying. Shortly after being given the memorandum, Glenn was confronted by all of the Argonne experimenters at once, and was taken completely by surprise. He told the people that we had come to the same conclusion that they had with respect to element 99, explaining how we had been misled by SF activity tailing from the nearby californium peak. He phoned the disturbing news to me that same day.

"It was quite clear that, prior to our call, they had not investigated the transcurium region beyond plutonium at all. That was understandable since they had their hands already full with the lighter elements, working with the new, very heavy isotopes of plutonium with mass numbers 244–246 and americium daughters. Then, too, the lighter element activity came from β -emitters with which they

were familiar, whereas α -particle emission was predominant in the elements beyond curium and they were not very well equipped for this. We had assumed that they would be glad that we had taken on the task of looking at the heaviest elements since that was our specialty. That was not the reaction of Martin Studier, Sherman Fried, and their colleagues at Argonne, however. They felt that they had been scooped and that was hard for them to take. After all, Argonne was the lab that had discovered ^{244}Pu in the debris, the important achievement that had helped to open up this whole new ball game. They had been in a perfect position to find elements 99 and 100 all by themselves. It was probably especially hard for them to take because normally their job was the dull one of doing the routine task of analyzing the atomic debris for known radioactivities. Now something fundamentally new and exciting had happened that they should have discovered for themselves.

"Apparently they had heard somehow that we were going to look for heavier nuclides and decided that they would look also. They knew that they had the advantage of much more material and they should be able to overcome their initial setback. New people were pressed into duty — Paul Fields, Herb Diamond, A. Hirsch, J.F. Mech, Gray Pyle, John Huizenga, as well as Studier — to separate out a transplutonium fraction and analyze what was going on. They were not experienced in this business, though, never having run a citrate cation column, and certainly not as adept as Stan and Gary, so they had not yet seen the 6.6-MeV alphas. Four days after Glenn's phone call to Argonne they finally performed a cation column experiment of their own.

"Since this was the very first time that they had ever tried that type of chemistry their column ran poorly and gave only a very crude separation of the heavy elements. We do not really know why they decided that the 6.6-MeV α -activity must be due to element 99 not 100, as we had claimed erroneously in our first memo. Their deduction was correct, as it turned out, but it became clear to any experienced nuclear chemist that it could not have been based on their chemical evidence. We found out later, when we saw their

data, that the californium and 99 elution peaks were very broad; in fact, they overlapped so much that one could only say that these elements had been *fractionated* — they certainly were not *separated*. With the small amounts of activity that they had they would not have been able to tell the difference between elements 99 and 100 without an element 100 tracer. This is proven by the fact that they could not even detect that the 6.6-MeV alphas from $^{253}\text{99}$ at the peak of the element 99 elution curve were decaying. This was because they grew in from ^{253}Cf almost as fast as they decayed, a consequence of the very poor separation of the two elements.

"Meanwhile, back in Berkeley, when we examined our own data we decided that the Argonne "6.35 MeV" activity could not be real since it was not observable in our data. We decided that our next objective must be to find a real element 100. It seemed quite possible to us that it could be kept alive in the element 99 fraction by a long-lived β -emitter, so the first step in the process of finding element 100 was to get more debris.

"January 10, 1953. Additional B-29 filters were procured as well as some "fallout" material from Louie Werner of the Naval Radiological Defense Laboratory nearby in San Francisco. (Werner was a former member of Seaborg's "Met Lab" team in Chicago. He and Burris Cunningham had first isolated microgram amounts of plutonium by their innovative ultramicrochemical techniques in 1942; see Chapter 3.) The new material was immediately dissolved and put through the chemical procedures.

"January 15, 1953. Another cation exchange resin column was being run and once again we were delighted to see something new — this time element 100! The activity had an α -energy of 7.1 MeV and a half-life of only about a day. It turned out that it was being kept alive by an element 99 β -emitter with a half-life of about a month. We were able to observe, after separation, the decay of the alphas in the element 100 fraction and their growth into the element 99 fraction and from the yield we decided that the mass number was probably 255. We learned later that the Los Alamos people had seen 7.1-MeV alphas a little earlier in a transplutonium fraction but did

not know where they were coming from and had not told anyone. They had feared that these alphas arose from some sort of background.

"February 2, 1953. Now that we had found a real element 100 we decided that we were tired of playing games with the Argonne crowd. We felt that we could trust Rod Spence, so we decided to ask him to be a kind of referee and he was told in detail of our results. (Spence was a very popular and unassuming person, so much so that he denied that Los Alamos was any more than a helper in the research on the Mike debris. As far as he was concerned, his group was just helping the heavyweights, Berkeley and Argonne. Later, when it came time to publish the results and the discussions were taking place as to just who would be on the paper or papers, he remained in the background trying to stay out of the controversy that had developed between Berkeley and Argonne. It was at Berkeley's insistence that Los Alamos was made an equal partner in the first publication of the Mike results.) Glenn then called Manning to exchange information in general and in that conversation he casually disclosed to him that we had just found an element 100 activity. He did not tell him any of the details except that it was definitely not an α -emitter of 6.35-MeV α -particles. He went on to suggest that Spence be the "clearing house" for any information on element 100 so that we could avoid the communication problems that we had run into with element 99. This stratagem worked very well and Argonne did not learn of our results on element 100 until Glenn reported them a month later at the February 18 Boner Panel meeting.

"Now Argonne was on the spot! They knew that Berkeley had found element 100 — what did they have to offer? They went back and scanned their pulse analyses for some wayward α -peak that they had overlooked. Even though they had much more material than Berkeley had it was difficult for them because their elution peaks were very broad and the element 100 activity must have been spread over a number of plates. Ideally, these should have been counted simultaneously to get the maximum sensitivity but with only one

pulse analyzer this was not possible. This meant interminable counting times. The result was that the efficiency for them to detect element 100 was much less than ours and they failed to see the 7.1-MeV alphas at all even though at this time their abundance was about 2–3% of that of the 6.6-MeV activity.

"February 4, 1953. Their situation now was desperate and we heard nothing from them for some time. Finally, in a phone call from Manning to Seaborg the news was passed on that they had found something but that they wanted to follow its decay for 10 days and thus would not make the agreed-upon deadline that had been set informally with Spence for publication in his secret overall report called Ivy. Rod thought that they were still pushing the idea of a long-lived element 100 with an energy of 6.35–6.4 MeV.

"February 18, 1953. But that is not what surfaced at the Boner Panel meeting in Washington. The Panel was concerned with the Long Range Detection of Weapons Tests, both domestic and foreign, and met occasionally to digest new results from the various bodies that dealt with these matters. Seaborg was a member of this body. The Panel was particularly interested in what Mike had produced, since they had heard that it was highly unusual and Glenn was scheduled to speak about the UCRL results. The group from Argonne, invited to talk about their results, included Manning, Fried, and Gray Pyle. The confrontation was bound to be dramatic because the two sides had such disparate results.

"Manning spoke first. He reported that Argonne had observed that the californium fraction exhibited 6.1-MeV alphas and SFs. He said that they had chemically identified an α -activity probably due to $^{253}_{99}$ with a half-life greater than 50 days and an α -energy of 6.5 MeV. The big surprise was something that Glenn had not expected. Argonne had now radically changed its stance. Manning claimed that they had chemically identified an approximately 15-day, 6.6-MeV α -emitter that came from element 100! Manning showed severely overlapping elution curves for elements 98, 99, and 100 with roughly equal intensities and only one elution drop between peaks. What a change this was! Argonne had flip-flopped and was now

claiming that there was a 6.6-MeV activity due to element 100, presumably with mass 254, just as we had erroneously done two months before!

"Seaborg talked next and reported that the UCRL group had also observed the 6.1-MeV californium alphas and, along with them, SFs which seemed to be decaying with a half-life of about two months. (This was 60.5-d ^{254}Cf .) Beyond that, though, the Berkeley results were substantially different. He reported that we had chemically identified a 22-d, 6.61-MeV isotope due to element 99 (thought to be of mass number 253) that was sustained by a ^{253}Cf β -emitting parent with a half-life of about 100 days (later found to be about 20 days). He then went on to describe the recent Berkeley chemical identification of an approximately 1-d α -activity from element 100 with an energy of 7.0–7.1 MeV (probably of mass number 255) that was kept alive by a long-lived β -emitting element 99 parent. This must have been a huge surprise to Argonne. It was not what they had expected at all!

"The Argonne team had fallen into a trap of its own making and manufactured an analysis that was completely wrong. Seaborg had told them that we had found some kind of element 100 activity without even hinting what it might be, so the Argonne group felt that it should exist in their data, too. In actual fact, though, it was not in their data! They had not yet seen the 7.1-MeV alphas, so did not realize what it was that we had detected. It is hard to understand why they did not detect these alphas; after all, Los Alamos had seen them. A possible reason for this is the following. Their pulse-height analyzer was the original one that Herman Robinson and I had built in Chicago in 1945. Since it had only 48 channels, these could have been biased so that the higher energy pulses from the 7.1-MeV alphas occurred off scale and thus were not seen at all! A good guess is that they probably had set the pulse-height analyzer so that they would have more energy resolution in the region below 6.6 MeV and this could have precluded their detecting alphas above maybe 6.9 MeV. Our analyzer at Berkeley was a duplicate of the one in Chicago but we never made that fatal mistake because of our

extensive experience in analyzing activities with high energies made in the 184-Inch Cyclotron bombardments in which we discovered the collateral α -emitting chains in the lead-uranium region.

"In any event Argonne knew that, regardless of its α -energy, element 100 would show up best on the leading edge of the elution curve for element 99. That was certainly true, but it was also true that here the $^{253}\text{99}$ was relatively free of its ^{253}Cf parent and it would decay with its normal 20.5-d half-life. On the other hand, at the peak of the elution curve for element 99 this was not the case. The californium peak overlapped it and many of the atoms of 99 which decayed would be replenished as the 17.8-d ^{253}Cf parent decayed. [Both of these $T_{1/2}$ values are the present day values.] Thus, for several weeks the activity of element 99 at the peak of their elution curve would show only a small decrease. The net result was that the decay rates of the 6.6-MeV alphas in these two regions of the elution curve behaved quite differently and convinced the Argonne group that they were observing two different activities, one due to element 100 and the other due to element 99. The slightly lower energy at the element 99 elution peak probably was due to the samples being a bit thicker for some reason.

"Later in the afternoon in a private conversation Seaborg vigorously tried to convince Manning, Fried, and Pyle of this interpretation but met with absolutely no success. They must have been in shock. Even two months later, as of April 17, 1953, they had not changed their minds about their interpretation and stuck by their claim that there was a 10-d 6.35-MeV element 100 α -activity.

"March 2, 1953. Meanwhile, back in Berkeley the work continued. Rod Spence had obtained large amounts of fallout from Mike by the radiation-hazardous operation of scooping coral from adjacent islands. Of course, this meant a much more elaborate chemical operation was necessary by Thompson's people because now, instead of just a foot-square piece of filter paper, the chemists had to dissolve as much as hundreds of pounds of coral. To handle the larger amount of inert material, a new facility had been established nearby in a small temporary building facetiously called the "ore house."

"Stan staffed this facility around the clock with students and one of them was Ken Hulet. He remembers fondly how Stan would set up a flow chart and they would follow it as best they could. It was no trivial task to handle a garbage can full of what was mostly calcium carbonate. It really was an exercise in chemical engineering. The basic chemistry that they used incorporated Thompson's favorite separation procedure, the basic bismuth phosphate process that he and Glenn had devised at the "Met Lab" a decade before. It was well worth the effort because it increased our stock of "goodies" by an order of magnitude. Hulet was a very unusual student. He had first come to the attention of Stan as a member of the Health Chemistry Group who not only monitored the radiochemical operations but also assisted in the operations themselves. Ken worked closely with Stan and Gary and earned their respect for his ability and judgment. They encouraged him to go on and try for an advanced degree, so it was no surprise to them that he later (1953) accomplished that as one of Seaborg's students. Two decades later he was a member of the team that discovered seaborgium (see Chapter 10) and was given the privilege of announcing to the world the decision to name element 106 after Seaborg.

"With this additional source of fallout the previous information was confirmed and extended. Now the half-lives became better known and interesting data on the other elements were obtained. A very important discovery that we made was finding a β -emitting isotope of berkelium, ^{249}Bk , with a 9-month half-life that decayed to 360-year ^{249}Cf . One or two decades later these nuclides would become the starting material for Berkeley when we began to produce isotopes of elements 104, 105, and 106 by heavy ion bombardment since it meant that pure californium targets with mass number 249 could be prepared from berkelium. More information was found on the properties of other isotopes of californium with mass numbers 254, 252, and 250 and this would lead us to discover the very important subshell at 152 neutrons.

"Argonne pursued the same research tactics and eventually found for themselves that their early data were grossly wrong in many

important ways. The result was that they began a process of revisionism. Now they too reported that the $^{253}_{99}\text{Cf}$ was kept alive by ^{253}Cf , a fact which they had bitterly disputed before. Now that they had finally seen the 7.1-MeV alphas they made the unbelievable claim that they had been seeing them all along! (Spence told Glenn that Manning wanted to insert into their part of the Ivy report that they had "seen the 7.1-MeV alphas repeatedly.") Although they tried to change the record to reflect their newly acquired information Rod Spence would not allow it since it had been agreed to have the Ivy report confine the input from the three laboratories to what was known as of March 20. Charlie Browne, who was weaving together an omnibus report combining the work of the three laboratories, froze the data as of that date and the Report makes it quite clear what each laboratory's position was as of that point in time.

The following entries from Seaborg's *Journal* for 1953 [Ref. 6.3, pp. 242, 256] are representative of this period:

'Sept. 1, 1953. Considerable time today was spent discussing the proper response to Winston Manning's letter of August 11 with Stan, Al, and Gary Higgins; we also talked about the current status of the debris program. Then, at about 4 p.m., I telephoned Rod Spence. I told him about the status of the 3000 pounds of debris, which has four to five times the activity of the previous batch and for which the chemical separation will be finished in a week or two. I asked if we could keep the entire element 100 fraction to look at the spontaneous fission activity before sending them their one-third. Spence agreed to this proposal. He told me that the Mike report is in press and that he will ask Smith (Ralph Carlisle) about declassification of the Radiation Laboratory version. After reviewing the difficulties in publishing together with Argonne (especially on element 100), I asked if Los Alamos would like to go in with Berkeley on open publication. Spence said that Louise Smith and Jack Barnes are working with Charles Browne. I mentioned all three, with Spence, as possible coauthors. Rod said he will talk it over with Browne and call back, possibly by Friday.'

'Sept. 16, 1953. ... Afterwards (10:50 a.m.) I telephoned Rod Spence in Los Alamos. Spence told me that he, Louise Smith, and Charlie Browne had discussed the matter of coauthorship on the initial open publication on elements 99 and 100 (as I offered to him in my call on September 1). Spence said they have decided against the idea on the basis that they feel they haven't contributed enough to deserve it; however, I urged them to reconsider and to discuss the matter with Norris Bradbury — I said that we will not consider the matter closed yet.'

"At this juncture we began to worry that other laboratories might discover lighter isotopes of elements 99 and 100 by the use of reactions with cyclotron-produced heavy ions. They would be able to publish that work without any problem and would feel that they should be able to name these elements. This might well happen before we could declassify the Mike work and it would make it difficult for us to claim priority in discovery. (Traditionally, the right to name a new element goes to the first to find it, but it is not clear that the world would accept that premise if the work is done secretly.) Bernard G. Harvey from Chalk River Laboratory had joined us by this time to work with Stan's group [Bernie was a world class nuclear chemist whom Stan and I had worked with occasionally in collaborative experiments; with great foresight, Seaborg had managed to wean him away from Canada]. Now we could expand our efforts substantially and since we had developed the cyclotron techniques to do heavy ion experiments, we decided that we should try them at Berkeley.

"November, 1953. The first project was to bombard ^{238}U with ^{14}N ions to try and find an α -emitting isotope of element 99. We had already discovered that californium metal could be vaporized out of a uranium metal target that had been bombarded by ^{12}C ions in the 60-Inch Cyclotron simply by melting it in vacuum, so we decided that element 99 should act similarly. (My colleagues used to kid me by calling this "blowtorch" chemistry!) This was done and, along with known isotopes of californium, a new α -emitter with a $T_{1/2}$ of

about 7 min was found. We thought that it was probably due to $^{247}_{99}$. The next step was to find out what its atomic number was. Could we do the chemistry fast enough to accomplish this? To prove this specifically we had to use a cation exchange column, so another step was added. The vaporized material was dissolved and loaded directly onto an ion exchange column. This was successful and we were able to prove chemically that the 7-min isotope was indeed due to element 99.^{6,4}

Quoting from Seaborg's *Journal* for 1953 [Ref. 6.3, p. 320]:

'Nov. 25, 1953. About 4 p.m. I telephoned Rod Spence to tell him that some of our fellows had definitely identified an element 99 isotope from $U^{238} + N^{14}$, declassified it, and sent a note to The Physical Review. I said the article was played down, definitely not a discovery paper with no name for element 99 suggested, and that the authors are not the same as the discovery authors. I cited the precedent in the case of americium. Spence agreed with the procedure and thought that this would react to speed up the possibility of the declassification of the discovery work. He said that he will talk with Ralph Carlisle Smith about it. I mentioned that Beckerley was now anxious to proceed with the declassification of the discovery work.'

Shortly thereafter, when this work was published in 1954 by Ghiorso, Rossi, Harvey, and Thompson,^{6,4} it was carefully pointed out that it did not constitute the discovery of element 99 because there was prior unpublished research on element 99 by the University of California Radiation Laboratory, Argonne National Laboratory, and Los Alamos Scientific Laboratory that had not yet been declassified.

"January, 1954. At about this same time the group began also to examine the products of the neutron bombardments of ^{239}Pu which had been going on for a long time at the MTR. As expected, we very quickly found large amounts of ^{241}Am , ^{243}Am , ^{242}Cm , and ^{244}Cm . In addition, since we knew what to look for and how to look for it,

we found ^{249}Bk , ^{249}Cf , ^{250}Cf , ^{252}Cf and the 6.6-MeV alpha particles of $^{253}\text{99}$. The next step was to make a new isotope of element 100 and with our new material this was straightforward. We had merely to bombard our new $^{253}\text{99}$ with neutrons at the MTR and look for a short-lived $^{254}\text{99}$ which should β -decay to $^{254}\text{100}$. This was done and soon we had shown that we could produce a 3.2-hour 7.22-MeV α -emitter that could be ascribed to $^{254}\text{100}$ sustained by its parent, the 1.5-day β -emitter, $^{254}\text{99}$. This work could be published readily since it was not secret and when it was published by Thompson *et al.*,^{6,5} and Harvey *et al.*,^{6,6} they were careful to again disavow that this constituted discovery of elements 99 and 100. This was a close call because a few months after this work was brought to the attention of the scientific public Manne Siegbahn, in a letter to Seaborg, mentioned that Atterling, Forsling, Holm, Melander, and Åström "had good indications that they had produced element 100 by bombardments of ^{238}U by ^{16}O in their big cyclotron." He went on to suggest that element 100 be named 'nobelium'! The publication of our work on 99 and 100 by these nonsecret methods forestalled claims by others to naming of the two new elements. Consequently, when I announced our choices for the two names, einsteinium for element 99 and fermium for element 100, at the first Atoms for Peace Conference in Geneva in August 1955, they were acclaimed without dissent.

"June 3, 1955. But before this announcement could happen we had to settle the matter of publication of the discoveries of these two elements. Seaborg and I met with Manning, Studier, Fields, and Fried at Chicago to discuss how we should proceed. In an attempt to reach some sort of agreement we debated at length the relative merits of publishing separately as Berkeley was advocating as opposed to publishing together as Argonne wished. I felt strongly that Argonne should not be a codiscoverer of element 100 but conceded that for element 99 this was reasonable. I felt that a fair compromise would be for Berkeley and Argonne to jointly publish the work on

element 99 and for Berkeley and Los Alamos to jointly publish the work on element 100. Argonne was not at all receptive to that idea. Details of the events of the last two-and-a-half years were gone over and over from the points of view of both laboratories *ad nauseam*. It was a very wearing encounter to everyone. Finally, it became clear to Glenn and me that Argonne was not willing to accept any reasonable compromise. They wanted to be codiscoverers of both of the elements no matter what the price!

"Glenn was desperate and felt that we had to emerge with some sort of agreement because he felt, and I must admit rightly so, that if we did not do so the whole disagreement would spill out into the public arena and result in a continuing brawl that would do no one any good. Finally, during the last intermission of our discussions he came up with an idea that struck me as brilliant. I felt that it would accomplish what I had been striving for and yet would allow a generous share of the credit to Argonne. But we were not certain that they would agree to it. His suggestion was that we would agree to publish the discoveries of both elements jointly if they would allow us to write the paper. All three laboratories would publish the Mike work as a joint report. To our surprise, the Manning group immediately agreed to this and Glenn and I withdrew to a quiet place and set ourselves to work writing the *Physical Review* Letter. In it we made it very clear that both elements 99 and 100 were discovered at Berkeley first. The Berkeley group would be senior authors and the Los Alamos group would be included.

"The Letter thus became a simple factual account^{6,7} of a remarkable episode in science. We were not sure that the Argonne group would accept it because it was so straightforward but we presented it to them for their approval. Somewhat to our surprise they accepted it without any changes. The following is the description of this meeting from Seaborg's *Journal*:^{6,8}

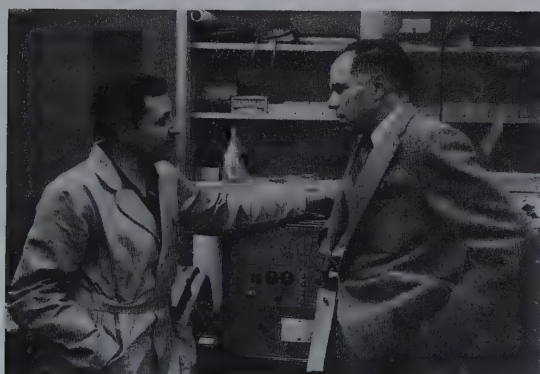
'In Chicago. Al Ghiorso and I spent the day at Argonne in discussions with Winston Manning, Marty Studier, Paul Fields, and Sherman Fried to try to come to agreement as to how to

publish our competing claims to the discovery of elements 99 and 100. We talked about the relative merits of publishing separately, as we (the Berkeley group) have been advocating if they continue to refuse to recognize our codiscovery of element 99 and sole discovery of element 100, or publishing together, as they (the Argonne group) want, conceding to them unjustified codiscovery of both elements. I finally decided, and convinced Al with some difficulty, that there is no way of publishing separately in an amicable and peaceful manner and that such a method of publication will inevitably result in a continuing public brawl. I then suggested that Al and I, using the name "einsteinium" for element 99 as preferred by the Los Alamos people, attempt to compose an article for joint publication to see if we could find agreement on it. We then immediately dictated to an Argonne secretary the document, entitled "The New Elements Einsteinium and Fermium, Atomic Numbers 99 and 100," which combined the salient parts of our two documents, UCRL-2981 ("The New Element Losalium, Atomic Number 99," by A. Ghiorso, S.G. Thompson, G.H. Higgins, and G.T. Seaborg) and UCRL-2947 ("The New Element Fermium, Atomic Number 100," by A. Ghiorso, S.G. Thompson, G.H. Higgins, and G.T. Seaborg). The write-up made it clear that both elements 99 and 100 were discovered at Berkeley first. This was satisfactory, without any changes, to the Argonne group, who also agreed that Ghiorso and the Berkeley group would be senior authors and the Los Alamos investigators would also be included as authors — thus the agreed-upon authorship was A. Ghiorso, S.G. Thompson, G.H. Higgins, G.T. Seaborg, M.H. Studier, P.R. Fields, S.M. Fried, H. Diamond, J.F. Mech, G.L. Pyle, J.R. Huizenga, A. Hirsch, W.M. Manning, C.I. Browne, H.L. Smith, and R.W. Spence.' Figure 6.7 shows some early photos of some of the discoverers."

"After this grueling day Winston Manning entertained the Argonne group and Glenn and me with a cocktail party at his home in Downer's Grove. As Glenn says in his *Journal*:^{6,8}



(a)

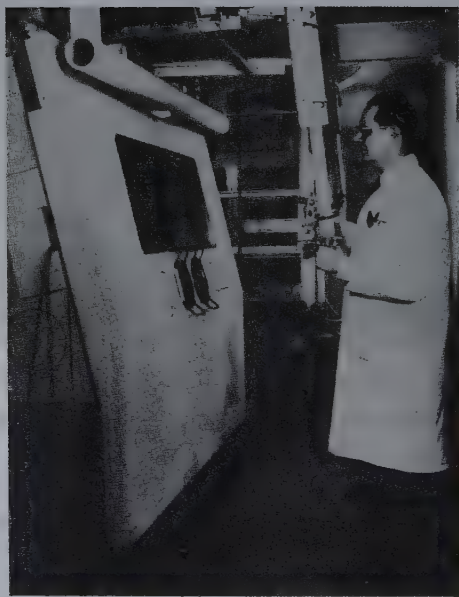


(b)

Fig. 6.7. Some early photos of the discoverers about the time when elements 99 and 100 were discovered. (a) Al Ghiorso and Glenn Seaborg with Dr. Earl Herald for his program *Science in Action* on KRON-TV, San Francisco. (b) Albert Ghiorso and Glenn T. Seaborg at about the time when elements 99 and 100 were discovered, 1954. (c) Charles Browne and Rod Spence in the early 1950s. (d) Paul Fields operating remote handling devices like those used in the isolation of einsteinium and fermium.



(c)



(d)

Fig. 6.7 (*continued*)

'Al compensated for his disappointment by enjoying an abundance of cocktails. He commented to me that he thought we should have taken our umbrellas on this trip à la Neville Chamberlain for this "peace in our time" conference — he also referred to it as a "Yalta" conference. Basically, however, he realizes that we did the right thing in negotiating this compromise and that the Argonne group made a far-reaching concession in allowing us to compose the document with its very favorable slant toward Berkeley.'

That was in the days when I used to drink! I didn't even notice the rough plane ride home."

6.3. Naming of Elements 99 and 100

The naming of the transuranium elements had first followed the system of using the planets of the earth as its model: thus Uranus, Neptune, and Pluto led to uranium, neptunium, and plutonium for elements 92, 93, and 94. Since there were no planets beyond Pluto, Seaborg resorted to using the analogous rare earth elements for the paradigm: and thus the sequence of europium (continent), gadolinium (scientist), terbium (city of Ytterby), and dysprosium ("difficult to get at") led to his suggesting for the homologous actinide rare earth elements the names americium, curium, berkelium, and californium (elements 95, 96, 97, and 98). The last is a "bit of a stretch" but Glenn defended it by saying that just as dysprosium was "difficult to get at" so was California difficult to get to in the early days of the West.

To have continued in this vein would have meant for elements 99 and 100 names analogous to holmium (city of Stockholm) and erbium (city of Ytterby). Ghiorso's favorite name had always been curium and he lobbied strongly that we should start a new paradigm of naming elements after famous scientists. The names of einsteinium after Albert Einstein (Fig. 6.8) and fermium after Enrico Fermi (Fig. 6.9), were natural standout selections that he and everyone else favored, so it was not hard to get agreement, strongly supported by Spence, to give the name einsteinium to element 99 and fermium to element 100. Once this had been decided Ghiorso decided that it

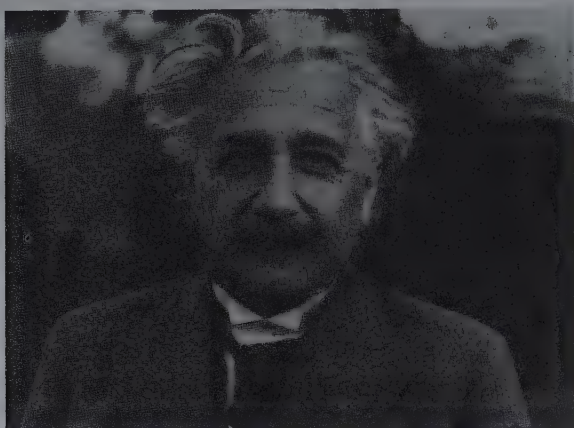


Fig. 6.8. Albert Einstein at UCLA, February 15, 1932.

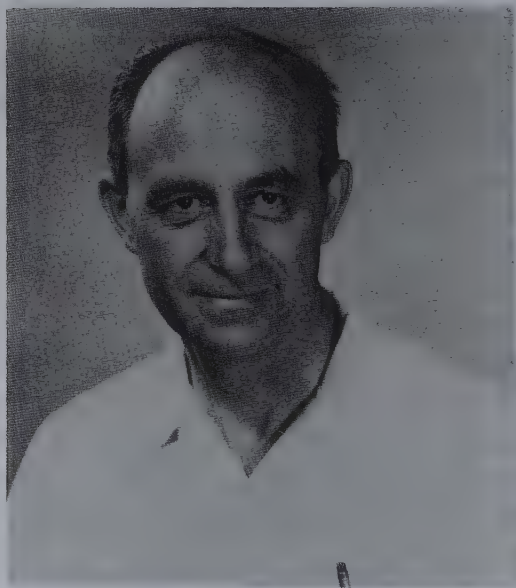


Fig. 6.9. Enrico Fermi, mid-1940s.

would be a nice gesture to tell Enrico Fermi of our decision while he was still alive; we knew that he would not live much longer because he was suffering from a terminal cancerous condition. He contacted Emilio Segrè to ask him to tell Enrico of our decision but found that he was not interested enough to do so — a not untypical Segrè reaction, Seaborg said! Five months after Fermi's death, Ghiorso wrote to Mrs. Enrico Fermi in April 1955, as follows: "I thought you might like to know that we are planning to name element 100 in honor of Enrico... It was my good fortune and privilege to know your husband in the days of the Metallurgical Laboratory project, and I can say from personal contact that science has lost a very warmhearted human being as well as its greatest physicist."

Other possibilities were considered for elements 99 and 100 before making the final decision for the names. The obvious name centurium for element 100 was suggested by many people. The possibility of naming element 99 or 100 after one of the laboratories was very seriously considered. It would have been natural to honor in this manner the Los Alamos Scientific Laboratory, which was responsible for staging the Mike explosion. In a phone conversation between Seaborg and Rod Spence, they discussed such possibilities as losalium, losalamium, losalamosium, and alamosium, and, in recognition of the acronym LASL, the names laslium, laslucium, and uclasium, the latter two in recognition of the role of UC (University of California) in operating the Los Alamos Scientific Laboratory. Another possibility, of course, was argonnium, after the Argonne National Laboratory. Berkeley had already been honored as the namesake for element 97. The scientists from the Argonne National Laboratory considered at one time the name phoenicium for element 99. Seaborg received one interesting letter suggesting arconium in honor of Arco, Idaho, for element 100. This correspondent pointed out that its lanthanide homolog, erbium, was also named after a town, Ytterby, Sweden. This suggestion was made after the publication of the research in which element 100 had been synthesized by neutron bombardment in the MTR at Arco, and before it was revealed that it had been synthesized earlier in the Mike explosion. This correspondent also

suggested that element 99 could be named after a location because its lanthanide homolog, holmium, derives its name from Stockholm; he suggested, therefore, that element 99 might be named ucalium, anlium, or losalium, after the University of California, ANL, or Los Alamos. It is clear that the manner in which erbium and holmium were given their names gave the discoverers the license to name elements 99 and 100 after a place had they chosen to do so.

The name and symbol athenium, Am, for element 99, and centurium, Ct, for element 100 appeared mysteriously in the literature in the early 1950s. It appears that this was an indirect result of a talk by Luis Alvarez at a conference at Oxford, England, in 1950, in which he outlined the future possibility for synthesizing these elements by certain nuclear reactions. This was misinterpreted as an announcement of discovery by the press and even reached some scientific journals. A consequence of this, apparently, was somehow the suggestion of the names athenium and centurium which had either a Spanish or a French source, and these names appeared in the Soviet literature for a while in the early 1950s. Soon after the letter to the editor in *The Physical Review* announcing the discovery of elements 99 and 100, another correspondent wrote as follows: "I stated very plainly, in diagram text and nomenclature, a new atomic theory which named element 99 ninety-nineum, symbol Nn, and element 100 centinium, symbol Ct. This is my creative knowledge, and I value and honor each atom ninety-nineum, 99, and centinium, 100, a million dollars each."

The announcement of the names einsteinium and fermium was first made at the Geneva International Conference on the Peaceful Use of Atomic Energy by Ghiorso just as the Letter to the Editor "New Elements Einsteinium and Fermium, Atomic Numbers 99 and 100" was appearing in the August 1, 1955, issue of *The Physical Review*.^{6,7}

6.4. Microscopic Quantities

It was not until 1961 that sufficient einsteinium had been produced through intense neutron bombardments of ^{239}Pu in the MTR to

permit separation of a macroscopic and weighable amount. B.B. Cunningham, J. Wallmann, L. Phillips, and R.C. Gatti,^{6,9} working at Berkeley on the submicrogram scale, were able to separate a small fraction of pure ^{253}Es . This was a remarkable feat since the total amount of material involved was only a few hundredths of a microgram of einsteinium. As was true with all other isolations of weighable quantities of the transuranium elements, it was possible to observe a macroscopic property — in this case, the magnetic susceptibility of einsteinium. A macroscopic property of the "metallic" zero-valent state of fermium, the magnetic moment, was measured in 1971 at the Argonne National Laboratory by L.S. Goodman, H. Diamond, H.E. Stanton, and M.S. Fried^{6,10} using the 3.2-h ^{254}Fm in a modified atomic beam magnetic resonance apparatus.

6.5. Publication

In the Spring of 1953, Browne wrote a description of the sequence of events that was ultimately accepted (with a few minor changes) by all of the parties involved. He combined the data from Berkeley Memo MB-IP-740 (March 19, 1953), Argonne Memo ANL-WMM-1076 Rev. (March 20, 1953), and Los Alamos data into a single classified report called Ivy Report WT-630 (October 1953). However, these data were not made public until August 1955 due to classification issues. And, in fact, these new elements were produced in a high flux reactor by slow neutron capture in plutonium, and the announcement of that work was made public prior to the publication in the August 1, 1955, issue of *The Physical Review* of the Letter to the Editor entitled "New Elements Einsteinium and Fermium, Atomic Numbers 99 and 100."^{6,7}

In that article the full story of the original discoveries in the winter of 1952–53 was told. It should be pointed out that the primary acknowledgment in this *Phys. Rev.* paper is to the personnel of LASL for the design and construction of the thermonuclear (Mike) device, which gave rise to the extremely high neutron flux required for rapid capture of at least 17 neutrons in ^{238}U to produce ^{255}U , which then β -decayed to einsteinium and fermium.

6.6. Limits to Production

As it turns out, fermium is still the heaviest element which can be synthesized in high flux reactors using neutron capture reactions. In subsequent nuclear tests, attempts have been made to produce and identify elements beyond fermium, but the heaviest isotope produced that was identified in those experiments (see R. Hoff in Ref. 6.1) is ^{257}Fm , which has a half-life of 100 d. In the 1960s, much smaller nuclear devices than Mike (kilotons rather than megatons) were detonated underground at the Nevada Test Site, USA, to see if heavier fermium isotopes (and possibly heavier elements) could be produced. Although several other heavy nuclides, such as ^{232}Th , ^{237}Np , ^{242}Pu , and ^{243}Am , were investigated for use as target materials, none of these showed any significant improvement over ^{238}U even though they were exposed to similar or higher neutron fluxes. Hoff discussed this in considerable detail^{6.1} and concluded that the observed yields of heavy nuclides could be explained as arising solely from capture in ^{238}U . An odd-even effect in the measured mass-yield curves is observed to reverse at about $A=250$ and has been explained as a result of multiple captures in the odd- Z element protactinium, formed by the (n,p) reaction on ^{238}U . The experimental data can be fit rather well by a combination of the yield curves for these two elements which depend on the amount of ^{238}U surviving fast-neutron fission and the amount converted to ^{238}Pa . Hoff also pointed out that extrapolation of the production yields suggested that detectable amounts of nuclides with $A>257$ were produced, but no evidence for them could be found by the radiochemistry teams. However, some 10^{20} atoms of ^{250}Cm (6.7 atom % of the curium) and 6×10^{17} atoms of ^{257}Fm were produced in "Hutch," the most successful of these tests, but recovery from the test debris some 200 m underground proved unusually difficult and netted only of the order of 10^{-8} of the total yield. However, these isotopes were invaluable for use in subsequent studies of SF properties and the ^{250}Cm was used as a target for the production by n-capture of a new isotope, 17-min ^{251}Cm . Further recovery attempts were abandoned

and it was suggested that in the future tests might be conducted in a more tractable medium such as salt to make recovery of these exotic nuclides easier.

Unfortunately, such tests would violate the Comprehensive Test Ban Treaty and, as if that were not enough, the mining and chemical recovery of the products would be prohibitively expensive at this time. That is too bad, because from a physics and chemistry standpoint it would be interesting to pursue the idea.

6.7. Role of Spontaneous Fission

A SF "disaster" was later (1971) found by Hulet *et al.*^{6.11} to occur at ^{258}Fm , which decays by SF with a half-life of only 0.380 ms. In 1975, Hoffman *et al.*^{6.12} produced and identified the 1.5-s spontaneously fissioning isotope ^{259}Fm at the Los Alamos van de Graaff Facility (Fig. 6.10) using the (t,p) reaction on a target consisting of only a picogram of ^{257}Fm . The target material was obtained through the DOE's transplutonium production program at the Oak Ridge National Laboratory, a program initiated by Seaborg during his tenure as



Fig. 6.10. Darleane Hoffman, Jerry B. Wilhelmy, and Joseph Weber, at the Los Alamos van de Graaff facility with the apparatus used in the 1975 discovery of 1.5-s ^{259}Fm .

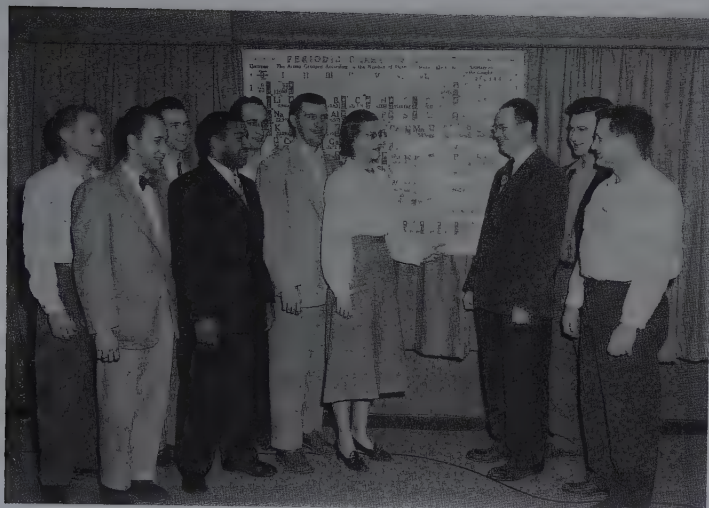
Chairman of the Atomic Energy Commission. Thus, it became clear that even if enough neutrons were captured to produce $^{258,259}\text{U}$, elements heavier than fermium could never be reached in this way because the decay chain would be terminated by fission at fermium, if not before; furthermore, ^{258}Fm is stable toward β -decay.

(a)



Fig. 6.11. Photos taken at the time of the January 23, 1978, Symposium Commemorating the 25th Anniversary of Elements 99 and 100, held at the Lawrence Berkeley Laboratory. (a) Some codiscoverers of elements 99 and 100 at the Symposium. *Front row:* Louise Smith, Sherman Fried, Gary Higgins. *Back row:* Al Ghiorso, Rod Spence, Glenn Seaborg, Paul Fields, John Huizenga. (b) Chemists who worked on elements 99 and 100 at the Argonne National Laboratory. *Left to right:* Lawrence B. Magnusson, Arnold M. Friedman, Charles M. Stevens, Philip A. Sellers, Herbert Diamond, John R. Huizenga, Gray Pyle, Paul R. Fields, Joseph F. Mech, Martin H. Studier. (c) Participants in the Symposium. *Left to right, bottom row:* Louise Smith, Darleane Hoffman, Kenneth Hulet. *Second row:* Sherman Fried, Gary Higgins. *Third row:* Al Ghiorso, Rod Spence, Don Ferguson, Norman Edelstein. *Fourth row:* Glenn Seaborg, Bill Carnall, John Conway, Paul Fields, John Huizenga, Dick Hoff.

(b)



(c)



Fig. 6.11 (*continued*)

Prior to this, from the separation of minute quantities of ^{257}Fm from subsequent "heavy element" tests, its SF properties were studied at Los Alamos and it was discovered that there was an enhanced yield of symmetric fission. This led to a renaissance of interest in the SF process, which has greatly expanded our knowledge of the fission process and our ability to model and predict the stability of new and still heavier isotopes and elements.

6.8. Commemorative Symposium

A symposium to commemorate the discovery of elements 99 and 100 was held in Berkeley on January 23, 1978,^{6,1} and in addition to reminiscences, many talks on the most recent research on einsteinium and fermium were presented. Some photos taken at that time are shown in Fig. 6.11 and include: (a) 8 of the 11 codiscoverers of einsteinium and fermium; (b) chemists who worked on the discoveries at the Argonne National Laboratory, and (c) a group photo of the participants in the symposium.

6.9. Outline of Important Points in the History of Elements 99 and 100^{6,13}

1. Detonation of the Mike device on November 1, 1952, on Elugelab Island in the Eniwetok Atoll in the Pacific by Los Alamos Scientific Laboratory.
2. Observation of "unusual" β -activity in the purified plutonium diagnostic samples at Los Alamos and Argonne due to $^{245,246}\text{Pu}$ and the subsequent discovery of the long-lived ^{244}Pu in mass-spectrometric measurements.
3. UCRL group decision (December 4, 1952) to look for trans-californium elements in Mike debris, made on basis of presence of ^{244}Pu in the plutonium fraction as established by Argonne group.
4. UCRL elution pulse analysis run No. 1 (Mike Citrate I, December 13–14, 1952). Filter paper source of material. Established presence

of 6.6-MeV source of alpha and SF activity which appeared to be due to transcalifornium isotope or isotopes.

5. UCRL elution pulse analysis run No. 2 (Mike TL Cit, December 16–17, 1952). Filter paper material. Proved 6.6 MeV due to transcalifornium isotope, as a result of its elution ahead of tracer ^{246}Cf .
6. Memorandum MB-IP-719 (dated December 19, 1952, written December 17–18, 1952) sent to Spence, Manning, AFOAT, Whitney, stated UCRL chemical identification proved 6.6-MeV α due to transcalifornium isotope, not sufficient to distinguish between elements 99 and 100. Suggested tentative assignment to $^{254}100$ on erroneous assumption SF due to element 100, as expected from systematics.
7. GTS phone calls to Spence and Manning (December 19, 1952). Told of discovery of transcalifornium activity, probably due to element 100.
8. UCRL elution pulse analysis run No. 3 (Mike TL Cit. 2, December 19–20, 1952). Material redissolved from run No. 2. Completely proved 6.6-MeV α due to element 99, assigned to $^{253}99$.
9. ANL elution pulse analysis run No. 1 (elution on December 23, 1952). Filter paper source of material. Independently established 6.6-MeV α due to element 99, assigned to $^{253}99$.
10. Memorandum ANL-WMM-1059 (dated January 6, 1953). Reported ANL chemical identification of 6.6-MeV α as element 99, possible 6.35-MeV α as element 100 (latter never found by UCRL to even a small fraction of ANL reported intensity).
11. UCRL elution pulse analysis run No. 4 (January 15–16, 1953). Larger filter paper source of material. Completely proved 7.1-MeV α due to element 100, assigned to $^{255}100$, with long-lived $^{255}99$ parent. Confirming run No. 5 (January 26, 1953) using material from redissolving run No. 4. The 7.1-MeV α -independently and previously observed, without chemical identification, by Los Alamos group.
12. GTS phone calls to Spence and Manning (February 2, 1953). Told Spence details of element 100 properties, told Manning only that discovery of element 100 had been made. (No word from

Manning to Spence on properties of possible ANL element 100 isotope.)

13. GTS and Manning report to Boner Panel in Washington (February 18, 1953). GTS reported ^{253}Cf β (≈ 100 d) \rightarrow 25399 (6.6-MeV α , ≈ 22 d); 25599 β -(long) \rightarrow 255100 (7.1-MeV α , ≈ 22 d); long-lived 6.1-MeV californium α , californium SF (≈ 60 days). Manning (WMM) reported ≈ 15 d, 6.6-MeV 255100 α , all of about equal intensities with severely overlapping elution curves, no report of β -emitting parents, reported SF due to californium. (GTS pointed out to WMM that he thought ANL " ≈ 15 d, 6.6-MeV 255100 " is actually ≈ 20 d, 6.6-MeV $^{25399}\alpha$, and hence they have reversed this assignment from element 99 to 100, and that the ANL > 50 d, 6.5-MeV 25399 decay sequence is complicated by incomplete californium–element 99 chemical separations.) The 6.6-MeV element 100 activity was never observed by UCRL group.
14. UCRL elution pulse analysis run No. 6 (GC1-C1, March 1, 1953), using 75 pounds of coral. Confirmed and extended previous work.
15. ANL elution pulse analysis run No. 2 (March 9, 1953), using 100-pound amounts of coral. Results interpreted much the same as previously.
16. Memorandum MB-IP-740 (dated March 19, 1953). Summarized UCRL data ^{253}Cf β (≈ 30 d) \rightarrow 25399 (6.6-MeV α , ≈ 22 d), 25599 β (≈ 30 d) \rightarrow 255100 (7.1-MeV α , 16 h), data on californium, berkelium, curium, and americium activities.
17. Memorandum ANL-WMM-1076 Rev. (dated March 20, 1953). Summarized ANL data, ≈ 15 d, 6.6-MeV 255100 α of much lower relative intensity than reported on February 18, 1953, 35–50 d, 6.6-MeV α , indefinite on parent ^{253}Cf but seemed to take issue with UCRL view that there is a long-lived ^{253}Cf parent. No "pulse-analysis elution" diagram for ≈ 15 d, 6.6-MeV 255100 α . Reported californium, possible berkelium, curium, americium, and plutonium activities.

18. Ivy report WT-630 (issued in October 1953). Data from MB-IP-740 and ANL-WMM-1076 Rev. and Los Alamos data combined into single report by Charles I. Browne.
19. ANL group again lowered intensity of their " ≈ 15 -d (now ≈ 10 -d), 6.6-MeV $^{255}_{100}$ " a relative to that of the " $35\text{--}50$ d, 6.6-MeV $^{253}_{99}$ " to ≈ 0.01 as of February 1953 and ≈ 0.1 as of December 23, 1953 (as a result of consideration of data taken during April and May of 1953). The UCRL group find that their data can establish even lower limits than these for this ratio as of those dates. Ratio of 0.1 as of December 23, 1953, corresponds, after correcting for decay, to about equal amounts of elements 99 and 100 "6.6-MeV alphas" at the time of the Mike shot, which could be easily checked in another similar explosion.

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Chapter 7

Mendelevium

7.1. Introduction

Mendelevium, element 101, was the first new transuranium element to be produced and identified “one atom at a time.” Albert Ghiorso, Bernard Harvey, Gregory Choppin, Stanley Thompson, and Glenn Seaborg announced its discovery, based on a grand total of only 17 atoms, in a short article published in 1955 in *The Physical Review*.^{7.1} It was synthesized via the $^{253}\text{Es}(\alpha, n) ^{256}101$ reaction in the 60-Inch Cyclotron at Berkeley. Especially noteworthy was that the target consisted of *only about 10^9 atoms* (< one-billionth of a milligram!) of highly radioactive 20-d ^{253}Es produced by irradiation of plutonium in the Materials Testing Reactor at the Arco Reactor Station in Idaho. The mendelevium was separated and chemically identified by its elution from a calibrated cation exchange resin column with α -hydroxyisobutyrate in the expected position for element 101. It was the last of the transuranium elements to be discovered and identified using direct radiochemical separation of the element itself. The discoverers suggested the name “mendelevium,” symbol Mv, in honor of Dimitri Mendeleev, the great Russian chemist who formulated and pioneered the use of the periodic table to predict chemical properties of unknown elements. This name was officially accepted later^{7.2} by the IUPAC, but with the symbol Md.

7.2. Tribute to Stanley G. Thompson

Stanley G. Thompson passed away on July 16, 1976. A tribute to Stan entitled “A Chemist’s Chemist” was written by Seaborg and

appeared in the Fall 1976 issue of the *LBL Newsmagazine*. It is shown in Fig. 7.1.

In March 1977, a symposium organized in his honor by the Division of Nuclear Chemistry and Technology was held during the 173rd American Chemical Society National Meeting in New Orleans, Louisiana. Session I, on "Chemical Properties of the Actinides," was chaired by O.L. Keller, Jr. Glenn T. Seaborg presented the first talk, on "Stanley G. Thompson — A Chemist's Chemist."

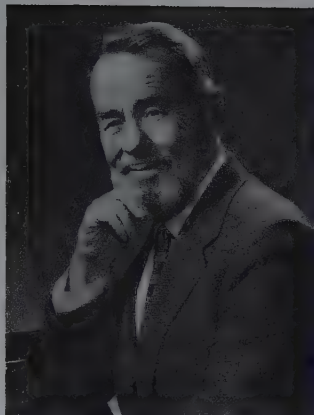
7.3. 25th Anniversary Symposium

A symposium commemorating the 25th anniversary of the discovery of mendelevium was held at the Lawrence Berkeley Laboratory on March 28, 1980. Of course, only four of the five discoverers gave accounts of the discovery. These reminiscences were followed by reports from E. Kenneth Hulet, Darleane C. Hoffman, and Arnold M. Friedman, summarizing the current state of knowledge about mendelevium as of the date of the symposium. Since the proceedings have only been published as an LBL report,^{7,3} we include here slightly edited, but essentially verbatim, extracts from the talks of the four discoverers present.

7.4. Introductory Remarks on the 1980 Symposium by Glenn T. Seaborg

I think it is appropriate to observe an occasion like the 25th anniversary of the discovery of a chemical element. I certainly think from a standpoint of history this is worth doing, but also, as has been the case for the other 25th anniversaries, it has resulted in a rather useful summarization of the information on the element as of the dates of the symposia. Mendelevium, I believe, is an interesting element, as you will learn from what you are going to hear today.

We have more information concerning the discoverers in this case; more on the record than we have had with any of the other elements. And I think you are going to be a little surprised in a few



A Chemist's Chemist

Throughout his career, Stanley Thompson was a chemist's chemist, "inventive and meticulous in his work, always seeking new fundamental knowledge. He avoided the administrative route to fame, preferring to work in the laboratory, and the result was an impressive record of discovery and help to the careers of his co-workers.

Stan's death this summer following a long illness brought a widespread feeling of loss at LBL, where he had worked for 29 years. He was known throughout the world as an outstanding pioneer and innovator in nuclear chemistry, a scientist who made crucial contributions during World War II, and an internationalist whose laboratory served as a home to scientist from many countries.

He retired from LBL in 1975. A native of Los Angeles, Stan obtained his A. B. in chemistry from UCLA in 1934 and his Ph.D. from Berkeley in 1948.

His first position was as a chemist in Richmond Laboratory of the Standard Oil Company of California. He left this position to join the chemistry Section at the University of Chicago Metallurgical Laboratory in 1942. There he conceived and directed the development of chemical process which was used for the extraction and isolation of plutonium from the production reactors that were built at Hanford in the State of Washington. In the course of this very successful development, hundreds of chemist were trained and worked under his direction.

He joined the Radiation Laboratory (as it was called then) in 1946 after the war and completed his Ph.D. thesis in a two-year period. Soon after this he led a research team on the synthesis and identification of berkelium and californium, the transuranium elements with the atomic numbers 97 and 98; a day-long program commemorating the 25th anniversary of this event was held at LBL just a year ago. He also participated in 1985 with the late Burris B. Cunningham in the first isolation of the elements in weighable quantities.

Stan Thompson was also a leader of the research teams that discovered the next three transuranium elements, einsteinium, fermium, and mendelevium (atomic numbers 99, 100, and 101). The discovery of mendelevium was notable in that this was the first element to be synthesized and identified on a one-atom-at-a-time basis.

He and his co-worker did extensive experimental work on the nuclear fission process, often exploiting the spontaneous fission of californium-252 for this purpose. His most recent work was in the field of heavy-ion reactions using the SuperHILAC. The work of the Thompson-Moretto group contributed substantially to the understanding of the various reaction mechanisms, especially the "relaxation phenomena" that take place in the interesting new inelastic processes.

Stan's wife Alice resides in Berkeley, and they have two married daughters, Ruth Ann Lincoln of Los Angeles and Joyce McClenney of Monterey, and two grandchildren. His mother and step father, Bessie and C. P. Fay, live in Valencia, California.

A Stanley G. Thompson Memorial Fund planned for undergraduate chemistry scholarships has been established through the University of California, Berkeley Foundation.

LBL Newsmagazine, Fall 1976

Fig. 7.1. "A Chemist's Chemist," by Glenn T. Seaborg, *LBL Newsmagazine* 1, 23 (1976).

moments as to what we have available to show you. It is very pleasant to have this reunion of the discoverers [Fig. 7.2] and their friends. A sad note, of course, is that Stan Thompson is not here. We certainly miss him and he was one of the key contributors to the discovery of this element. As you know, as I have stated on a number of occasions, Stan was a lifelong friend of mine; we knew each other when we were 13 years old, freshmen starting high school in the Los Angeles area and, as fate would have it, we spent a good deal of our life collaborating on research in a number of areas.



Fig. 7.2. Four of the five codiscoverers of mendelevium at the Lawrence Berkeley Laboratory on March 28, 1980. *Left to right:* Gregory R. Choppin, Glenn T. Seaborg, Bernard G. Harvey, and Albert Ghiorso. The fifth codiscoverer, Stanley G. Thompson, had died by this time.

Now, I am going to lay the groundwork with some descriptive remarks concerning the background for the discovery of this element illustrated by a number of slides. This element was really the turning point in the synthesis of new elements. This is the first element whose discovery was based on the one-atom-at-a-time approach, and the first element that used some of the techniques that are necessary for detecting isotopes or elements on such a small scale; all of the elements that have been synthesized and identified since mendelevium have used basically the techniques that were worked out for this occasion.

I am going to be rather brief, because I am sure that the reminiscers who are following me are going to fill in some of the details. The reaction ${}_{99}^{253}\text{Es} + {}_2^4\text{He} \rightarrow {}_{101}^{256} + n$ was used for the mendelevium discovery experiment. It was remarkable, in that this was really the first time that such a small amount of target material was used, an invisible amount, and I mean a really invisible amount, something of the order of 10^9 or 10^{10} atoms. And the einsteinium-253 had been synthesized over a number of years by the bombardment of lighter isotopes, beginning with plutonium in the Materials Testing Reactor at the Arco Reactor Station. This was then put on a gold backing foil and bombarded in the 60-Inch Cyclotron with helium ions (40 MeV), with the expectation that this reaction might occur, based on previous experience with reactions of this kind.

A calculation was made before the experiment was attempted to indicate whether it might be feasible to produce and identify such an element. The following rough calculation shows that the number of atoms that might be produced would be approximately equal to the number of atoms of target material times its cross section times the ion beam intensity times the time of bombardment, which, of course, would be related to the half-life of the product when bombarding for a time of the order of its half-life. On that basis, using for example 10^9 atoms, an estimated cross section of about a millibarn, based on measurements of similar reactions in this region, and a beam of 10^{14} helium ions per second (which was only obtained by literally rebuilding the 60-Inch Cyclotron because we weren't getting beams of

this order of magnitude), and an irradiation time of the order of 10^4 seconds, we came out with the result that we would expect perhaps one atom per experiment.

$$N \cong N_t \sigma \cong \text{It}(10^9)(10^{-27})(10^{14})(10^4) \cong \text{one atom per experiment.}$$

And it was on this basis that it was decided that it was feasible to go ahead.

Now one of the key changes or improvements in the technique that was introduced here for the first time by Albert Ghiorso, was the recoil technique, placing the target element on the opposite side of the target from the beam and catching the recoiling atoms on a catcher foil. Some coworkers had already done this in a few previous experiments, I believe, at the 184-Inch Cyclotron. This was then applied for the first time to the identification of a new element in this manner. The identification was made as shown here in a schematic manner [Fig. 7.3] in a Russian publication called *Priroda* (I believe that means "Nature"), which appeared shortly after the announcement in *The Physical Review*^{7.1} of the discovery of element 101. In this case, the helium ions, labeled 1, struck the target, gold and einsteinium-253 on the back side of the gold foil, labeled 2, and then, the gold catcher foil, labeled 3, was symbolically dissolved in the crucible shown, labeled 4. The transmutation products were then put through a Dowex-50 ion exchange column, labeled 5, actually at high

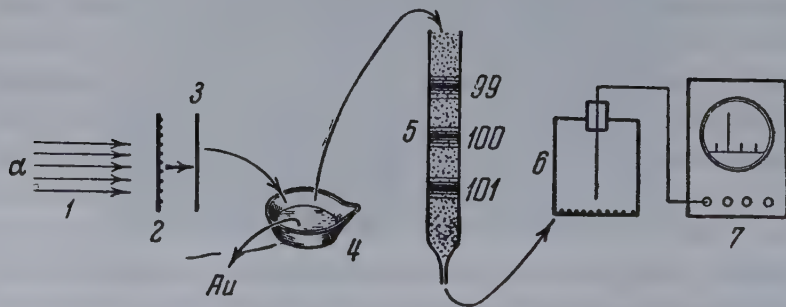


Fig. 7.3. Schematic procedure published in about 1955 in the Russian publication *Priroda*.

temperature (87°C), and eluted with an α -hydroxyisobutyrate solution, and then symbolically detected, as shown here in an ionization chamber, labeled 6 and 7, to measure, in this case, spontaneous fission (SF).

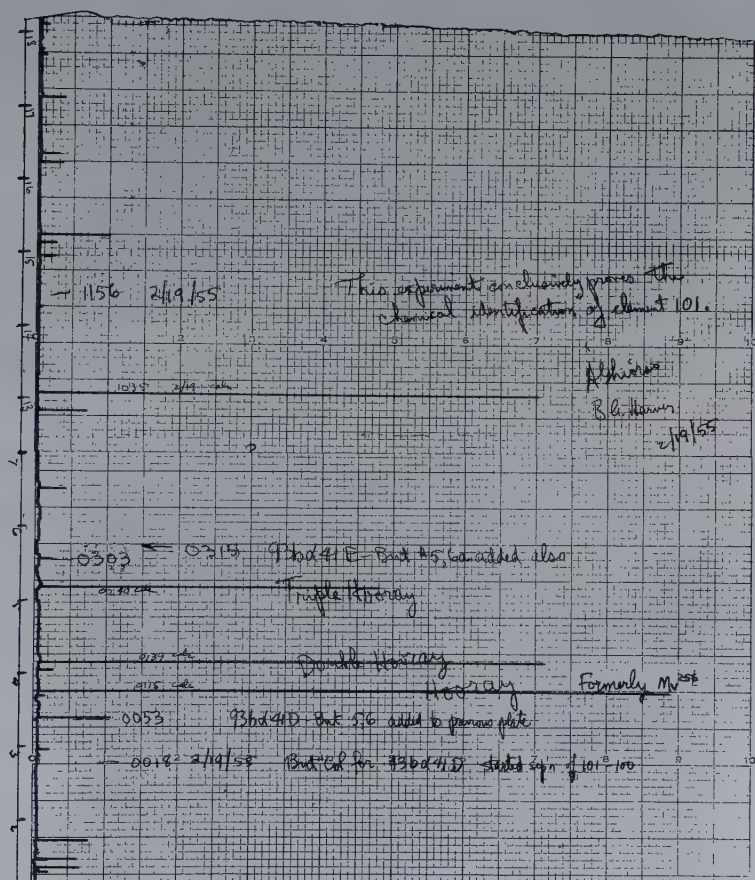


Fig. 7.4. The ionization recording chart showing the first four events of the disintegration of mendelevium. The ordinate is the event time while the abscissa is the intensity of the ionization. The four pulses occurred at 1:15 a.m., 1:37 a.m., 2:40 a.m., and 10:35 a.m. on February 19, 1955. At 11:56 a.m., Ghiorso and Harvey made a note directly on the chart: "This experiment conclusively proves the chemical identification of element 101."

Next we see the famous picture [Fig. 7.4] of the recording of their data, with the handwriting of Ghiorso and Harvey made during the all-night experiment when the discovery was made. As work went on it appeared that there was an SF activity that was involved in a kind of complicated double decay, the mendelevium-256 actually decaying by electron capture with the half-life indicated at that time to be a little under an hour, now known to be 76 minutes, to an SF activity, fermium-256, decaying with a half-life of 2.6 hours. The mendelevium was isolated, in the manner that I indicated [Fig. 7.3], by the ion exchange adsorption-elution technique. Then, in the definitive experiment, the SFs were recorded in this manner during the night of February 19, 1955. The first one was identified with a "Hooray, formerly Mv^{256} ," then a "Double Hooray" and a "Triple Hooray"; then there was a fourth one, and the inscription "This experiment conclusively proves the chemical identification of element 101," signed by A. Ghiorso and B.G. Harvey, and dated February 19, 1955.

We have an advantage over the other 25th anniversaries, in that we have pictures taken at approximately this time, actually a little later in 1955. A movie of a re-enactment of the discovery was filmed as part of a TV program of ten shows on the chemical elements. This series included this episode in one of the later programs which was devoted to the heavier transuranium elements. Here we see, taken from this film, a younger Albert Ghiorso and Bernard Harvey [Fig. 7.5]. And we see Stan Thompson [Fig. 7.6] at the other end carrying on the chemical identification.

As was often the case in the discovery of these elements, a party was held, in this case at Larry Blake's restaurant in Berkeley, celebrating the event. And here at the head table, we see Bernie Harvey, Albert Ghiorso, Bernie Rossi, me, and Nels Garden [Fig. 7.7]. Stan is not in these pictures because he was away in Sweden on sabbatical leave by the time we got around to this party. But here he is in effigy [Fig. 7.8].



Fig. 7.5. Al Ghiorso and B.G. Harvey; re-enactment of the element 101 discovery.



Fig. 7.6. Stan Thompson (*right*) and Bernie G. Harvey; re-enactment of the element 101 discovery.



Fig. 7.7. Mendelevium (Md) party at Larry Blake's restaurant, head table: Bernie G. Harvey, Al Ghiorso, Bernie Rossi, Glenn T. Seaborg, and Nelson Garden.



Fig. 7.8. Party at Larry Blake's; Glenn T. Seaborg and Al Ghiorso with an effigy of Stanley Thompson, who was in Sweden on sabbatical leave.

At a little later time, the *Daily Cal* ran a story that I happened to find in my files, and I am going to read some excerpts of this that I think you might find amusing. The story goes as follows:

"The University of California Nuclear Metaphysical Laboratories have announced a startling new finding in the world of atomics. The new discovery is an entirely novel element named 'percentium' by its discoverer, the 15½-year-old Leonardo da Vinci. The element, number 101 in the atomic series, follows element number 100, 'centium.' The youthful da Vinci said that this was the reason for the new element's name, percentium. 'The interesting fact about percentium,' said Moosbrugger, 'is that it has a negative half-life. That is,' he went on, 'its radioactivity and total mass increase 1 percent every 100 years. Probably it is the first of a series of elements that spontaneously integrates the successive members of the series.'

"'Percentium is characterized,' said M. Morris, a lab assistant on the project, 'by a satellite proton revolving around the nucleus inside the electron sheath. Preliminary investigations indicate that all the higher elements beginning with number 101 have similar binary nuclei. The satellite proton of number 101, it is believed by the department, picks up neutrons and protons from surrounding matter.'

"The process is presumed to continue up to element number 202; at this point the twin nuclei are of equal mass. When number 202 adds another proton to the satellite, the latter becomes more massive than the primary nucleus, and the resulting short-lived atom of element number 203 undergoes fission with emission of a proton and a neutron, and becomes two atoms of number 101.

"Apparently natural processes are incapable of producing any of the elements beyond number 100. If they were so capable, the universe would be overflowing with percentium. It would appear that the artificial production of percentium has initiated a slow cosmic revolution in the

atomic population. It is estimated that after 101 billion years all the matter in the continuum will consist of the elements from 101 to 203 inclusive.

“‘But this is nothing to worry about,’ said Kidder with a grin; ‘we’ll all be dead by then.’”

And, by the way, I would also like to call your attention to the fact that in 1972, in a journal that came out periodically, *Adventures in Experimental Physics*,^{7,4} edited by Bogdan Maglich, we have a rather good description by Al Ghiorso of the discovery of element 101.

[Seaborg then showed a film of the discovery experiment recreated by the discoverers a few months later. After the film he made the following comments.]

I am not sure this film was received with the respect that it deserves. Actually, I do not think that the predictions I made at that time were so bad. You must recall that we had not begun to use heavy ions yet, and, of course, everything beyond element 101 was out in the unknown. I do hope that some of the young actinide chemists who are present in the audience this afternoon got some tips on how to carry on their work in this region. Yes, I think there were some practices there that wouldn’t be allowed today, even to the extent perhaps of not being allowed to show the samples of actinide elements as I did on educational TV. I noticed, however, that I didn’t have plutonium among those that I demonstrated. I do not recall whether there was a prohibition against that even at that time.

7.5. Reminiscences from the 1980 Symposium

7.5.1. Albert Ghiorso

7.5.1.1. Introduction: Dr. Seaborg

Now we get on to the first of those among us who are going to reminisce a bit today, and that is Albert Ghiorso. I am going to dispense with any long introduction. We all know Albert. He began his career as a nuclear chemist at the Metallurgical Laboratory of the

University of Chicago in 1942. It was there, after carrying out some of the essential war work, that he could go on to broader investigations that led to the discovery of the next two transuranium elements, elements 95 and 96, americium and curium. He returned to the University of California, the Radiation Laboratory here on the hill in 1946 and in the intervening years has been the key person in the synthesis and identification, i.e. the discovery, of all of the heavier transuranium elements, up to and including element 106.

7.5.1.2. *Reminiscences: Dr. Ghiorso*

In thinking about what I should say on this 25th anniversary of the discovery of mendelevium, I decided that it might be of interest to tell the story of how the idea originated for this unique experiment — the bombardment of a nearly weightless target to make a new element, one atom at a time.

I used to find that when I went on trips I would inevitably get new ideas; this particular one occurred on a plane trip to Idaho. Bernie and Stan and Greg and I were going there this time to do some experiments with ^{253}Es to make new nuclides by neutron bombardment in the MTR high flux reactor. On the plane I started playing with some numbers related to the amount of einsteinium that we could obtain, and I suddenly realized that if we took the maximum amount of ^{253}Es that could be obtained at that time, about 3×10^{10} atoms, and bombarded it with a beam of 100 microamperes per cm^2 of ^4He ions, we would make one atom of element 101 every five minutes or so assuming a cross section of about a millibarn. The assumption of beam intensity was about an order of magnitude greater than had ever been obtained, but I blithely assumed that this problem could be overcome.

Very excitedly I took the calculations over to the others and we started to discuss the possibility of making the new element. I remember that Stan and Bernie were not enthusiastic. One of the most critical problems was that the einsteinium target had a half-life of only 20 days, so it would be necessary to do the experiments in

a very limited time. I argued that we could make it a little easier by making only one target, a recoil target. This would allow us to separate the transmutation products without dissolving the target by merely catching them in a foil placed next to it. This had never been done before with low energy light ions, but I was confident that the technique would work. We finally decided to make the try, but to reach our goal took a lot more effort than I had envisioned.

In principle, the idea was perfect, but it took us a while before we were able to make it work. We made something like five targets before we had a successful one. The reason for our difficulties was that we at first made the targets by vaporization (I was known as the blowtorch chemist in those days!) and yet they were too thick. We incorporated ^{244}Cm with the ^{253}Es so that we could monitor our success by the amount of ^{246}Cf that was caught in the recoil catchers. The reason that the early vaporized targets didn't work properly was that a thin transparent deposit of tantalum oxide or tungsten oxide was also vaporized onto the target and this absorbed the fusion recoils very readily. After a number of failures (after each failure the target material had to be recovered and repurified), Bernie got disgusted and said, "Let's electroplate the target." He did so and the target worked very well — an almost weightless target in a very small area. That target survived many bombardments but by the time that we got to the crucial experiments the amount of ^{253}Es was down to about 10^9 atoms, a very marginal amount.

Before we got to this point, however, we had to have a cyclotron that would give us a helium ion beam of at least 100 microamperes per cm^2 — this would be 5 microamperes through an area of 0.05 cm^2 . At that time (this was well before strong-focusing quadrupoles came into use) the only way we could get such an intensity was by taking advantage of the vertical focusing inherent in cyclotrons. We used the internal beam just after it was deflected. At this point the beam was only $1/32''$ high and $1/4''$ long, and this is where we placed the target. The target and recoil catcher cooling requirements as well as the intense radiation problem made the target probe equipment quite complicated. After each bombardment the catcher

foil became extremely radioactive, but by judicious quick manipulation we were able to keep our personal radiation exposures down to safe levels.

It was also necessary to improve the 60-Inch Cyclotron to make the experiment possible at all. This was done by Bernie Rossi, who is no longer with us, having died a few years after the experiments. I would like to pay tribute to him not only for his work in making this discovery possible but also for his role in the development of heavy ion technology. I used to work with him one day (and one night) a week to try and accelerate various heavy ions to usable energies and intensities. One of our important findings was the discovery of harmonic acceleration by which C^{2+} and C^{6+} could be accelerated simultaneously. We even developed a beryllium beam at that time! This heavy ion work led directly to the building of the HILAC. Rossi played a very crucial role.

The target problems were considerable but they were overcome. The trouble was that we didn't see anything — no short-lived alpha particles that could be identified as originating from element 101. The reason for the fast little car seen in the film (my supercharged Volkswagen) is that we were sold on the idea that the new hypothetical alpha activity would have a half-life of about five minutes so that it was necessary to cut the time from the end of each bombardment until the end of chemical separations to an absolute minimum. We used to telephone the guard at the Laboratory gate that we were coming so that there would be no delay in making entry. I remember one time, however, when someone didn't phone. As usual, as the driver, I raced through the gate going like mad. This particular guard, who was a real eager beaver, jumped out of his post and shouted, "Stop or I'll shoot!" Well, believe me, I wasn't about to stop — I thought he just *might* shoot! He was very much distressed; he came up to our lab afterwards and we apologized but said that we were too busy with the experiment to talk about the incident at the moment. We got away with it for a while.

As the experiments proceeded we became discouraged, because we couldn't find anything after using several days of bombardments.

One night, after our usual failures, we decided to look for something longer-lived. We made a somewhat longer bombardment around midnight and the separated transeinsteinium fraction was placed in a counter for an overnight alpha pulse analysis. I had hooked up a special circuit that would also record the pulse height of the "big kick" that would come from any SF that might occur. This circuit was connected to a large chart recorder and we left for the night.

In the morning a look at the chart showed two very high energy events that could only come from SFs, and they were separated by a few hours from one another and the end of the bombardment. Just these two events in all the bombardments that we had made — what did they mean? I was very bold and proposed that we had produced an electron-capturing isotope of element 101 decaying to element 100 which then underwent SF decay with a half-life of a few hours. The mass number would be 256.

This hypothesis completely changed our course of action. Up to this point we had assumed that the isotopes of element 101 that we would make would be short-lived α -emitters. There was no way of knowing that they would be highly hindered for α -decay. And now we had the possibility of finding an ec nuclide decaying to one undergoing SF with both nuclides having reasonable half-lives. This was a really wild lucky guess considering that our only evidence was the two "big kicks." Our strategy now was to assume that the $^{256}_{101}$ isotope had a half-life of about an hour and the $^{256}_{100}$ isotope had a half-life of several hours. A number of bombardments were then made and the successive element 101 fractions combined and placed in the alpha grid chamber so that we could analyze for high energy events. (In later experiments we used a group of counters that counted only fissions to obtain a complete chemical elution curve.) In the film, you saw a picture of the chart showing the times when we observed SF events in the element 101 chemical fraction. Our various "Hoorays!" written on the chart were deeply felt.

But it wasn't 3 a.m. yet. We had decided that if we found the element we would call Glenn at that time to let him know the good news! The last one came in at 3:15 a.m., so I called him around

3:30 a.m. Although quite sleepy, he, of course, was very pleased. I went one step further and told him that we had not only found the element, we had also decided on a name for it — “mendelevium.” The name suggestion came about a year before our discovery, when Jack Hollander proposed it at lunch one day. He said, “Why don’t you guys name an element after the famous Russian chemist Mendeleev?” We all thought it was a good suggestion but difficult to implement because of the Cold War, but filed it away in our minds for future consideration.

After we had finally found the recalcitrant element, we discussed among ourselves what its name should be and found that we were in complete agreement about using the name “mendelevium.” However, we felt that if we used the cautious approach and asked the opinions of Lawrence and Seaborg they might agree for scientific reasons but demur out of political considerations. Thus we felt that an aggressive approach might be in order — that if we just called it “mendelevium” maybe it would be all right. And it was. I think Glenn must have gulped over the phone but he didn’t object. Neither did anyone else! Everyone seemed to be pleased as a matter of fact. At the 1955 Atoms for Peace Conference in Geneva, the French chemist Haissinsky told me that our naming of element 101 in honor of a Russian scientist had probably done more good for international relations than anything that John Foster Dulles had ever done!

In subsequent trips to Russia, Glenn and I both found that the gesture was warmly remembered.

7.5.1.3. *Comments: Dr. Seaborg*

I think it is true that the naming of this element, mendelevium, had a dramatic impact in furthering, in a positive way, American–Soviet relations at that time. I think it really is rather difficult in today’s atmosphere to realize what a step that was at that time. It was quite a step, and there were a number of people who criticized us for it.

I recall that at the time of the visit of Vice-President Richard Nixon to the Soviet Union in 1959 — the time of his famous kitchen

debate with Premier Khrushchev — it occurred to me that it might be useful if Nixon knew the story of mendelevium. He happened to be a long-time friend. I therefore wrote him a letter and told him that we had recently named this element, mendelevium, after the Russian chemist hero, and that at some point or other that might be of some advantage to him in his discussions. I learned later that he used this information, i.e. he told some people about this and got quite a good reception. And one of the aftermaths of that was that a little later I got a copy of Mendeleev's chemistry book, autographed by Mendeleev, from a secondhand book dealer in the Soviet Union, who told me that he had learned about this from Vice-President Nixon. This was a book that the book-dealer had picked up somewhere; actually it was the book that was sent to a physician in England who played an important role in treating Mendeleev's son for some sickness at the time of a visit that Mendeleev made to England. So, that is an interesting aftermath.

7.5.2. Bernard G. Harvey

7.5.2.1. Introduction: Dr. Seaborg

The next codiscoverer we are going to hear from today is Bernard Harvey. I first learned of Bernie's existence when he was working at the Chalk River Laboratory. He was working on the chemistry of plutonium. And he had written a report. He, with a coworker or two, covered much of the work that the whole American team had done on the chemistry of plutonium. So I thought, This is certainly a remarkable chemist; I want to meet him. After I met him on one of my visits to Chalk River, I thought this would be a great fellow to bring to the Radiation Laboratory at Berkeley, and I communicated that to him. For a while, we had to overcome the prohibition on any foreigner — in those days even a Canadian was considered to be a foreigner — and it took some time, a number of years, before we got permission to offer him a position. And then, as Bernie has pointed out many times, I wrote him a carefully worded letter

offering him a temporary position in the chemistry group here at the Radiation Laboratory and that is the only thing he has to show for his present position. He is still here on a temporary status. This has never been rectified or amplified, and it is still the basis on which he is here.

Soon after he arrived, within a year to two, he got involved in this experiment on mendelevium that we are commemorating today. When the 88-Inch Cyclotron was built toward the end of the decade of the 1950s and the early 1960s, he took over as the Director and served in that capacity until last year, and now he is back at the 88-Inch Cyclotron. Bernie, could we hear from you as to what you remember about those days.

7.5.2.2. *Reminiscences: Dr. Harvey*

Thank you, Glenn. I have to correct one remark that you just made, though. It's true that when I came to the Lab in 1953 I had only a temporary appointment, and that remained true until about three weeks ago, when I received a letter from President David Saxon telling me that at last my appointment had been made permanent. I must say during those almost 27 years, the world's longest temporary appointment, I never really worried about when it was going to run out.

Well, I think the key to the successful experiment that you saw in the film and about which Albert and Glenn have talked was really technique. There were several new techniques that had to be developed and we were lucky (or perhaps we weren't lucky, we were smart) that they all happened to come together at the right time.

First of all, we had to make the recoil target. My memory, and perhaps Greg can correct me if I'm wrong, is that the electroplating technique that we used was developed by Alfred Chetham-Strode, who, alas, is no longer with us. Without that technique, as Albert said, the evaporation from a hot filament technique didn't work because of all other materials that were evaporated on the surface of the target. The electroplating method worked beautifully and it gave

a very high yield, which, of course, was very important for handling such a rare product as the einsteinium target material. So that was one technique that we just happened to have available to us at the crucial moment.

The other technique that we worked hard on, and that we also needed for this experiment, was the ion exchange separation on the Dowex 50 column. It was Greg's idea to use α -hydroxyisobutyric acid as the eluant. We tried many different eluants and we tried all kinds of conditions. I believe that Greg and I ran an average of three ion exchange columns a day, testing different techniques, over a period of many months in order to be sure that when we started a separation we would know exactly what was going to happen. In fact we knew to one drop where each element would appear. When I first came to Berkeley, there were frequent surprises where sometimes curium, for example, would come off at drop 150 and sometimes it would come off after about 150 liters of eluant. Such an uncertainty, of course, was quite impossible to live with when dealing with an experiment as difficult as the mendelevium experiment. But we really had that down cold; we refined every little detail of the technique of ion exchange columns, and as a result of that we were able to tell exactly where the interesting activities would appear. In fact we got so good at it that I remember working up one of these plutonium samples that had been in the Materials Testing Reactor, where one had many curies of fission products and curium and other heavier elements. After we ran the first ion exchange Dowex 50 column to separate out the uninteresting americium and curium from the interesting heavier things, we wanted to separate the really heavy elements one from the other in a second Dowex 50 column, but there was so little curium left after the first pass, we had to put some back in as a tracer so that we would know what was going on in our column. So, the key to it all was the ion exchange, the electroplating, and, of course, the development of the cyclotron to which Albert referred.

I think there was another very important factor in the success of this experiment. It was that the group of people working on it were

highly compatible one with the other and we all worked pretty hard. Albert made the remark that he's lost weight since those days and, seeing the film, I think I can say the same of myself. We needed it perhaps. But it was a good group, we were good friends, we worked very hard on this and many other experiments, and it was a lot of fun; it really was.

One final comment — I'm not sure I ever saw the film series on the television screen. The reason was that KQED, which was then a very new educational television station in the Bay Area, showed it at a most unfortunate time of day, and although my wife tried hard to get the children to watch their father, she was usually unsuccessful because the time of the film coincided with something more interesting, perhaps the *Lone Ranger*. Or perhaps it was at 6 a.m. — I don't remember. The children certainly had their priorities, which were not necessarily those of the makers of the film. So, Albert, Glenn, Greg, it was a lot of fun, and I must say I thoroughly enjoyed working with you. I wish we could do it again.

7.5.2.3. *Comments: Dr. Seaborg*

The TV show *Lone Ranger* was difficult to compete with. I remember once I appeared on the KQED program *Science in Action* as a guest. It was broadcast at some prime time like 7 p.m., and after the show was over I drove home. As I drove into my driveway, a number of kids in our neighborhood came running over and said, "Oh, Glenn, we just saw you on TV." I said, "Well, how did you like it?" They said, "Oh, we changed over to the *Lone Ranger*."

7.5.3. *Gregory F. Choppin*

7.5.3.1. *Introduction: Dr. Seaborg*

Next, I have the pleasure of introducing the discoverer of α -hydroxyisobutyrate as an eluting agent. This, by the way, is still the best eluting agent so far as I know for the separation of the actinides,

25 years later. I do not know why you don't get busy, Gregory, and develop something better.

Gregory Choppin joined our group in 1953, coming up from the University of Texas. He immediately began to work in the heavy isotope field with the heavy isotope people. As I recall, he was involved in the identification of the neutron-produced isotopes from the MTR at Arco, Idaho; isotopes like berkelium-249, californium-249, -250, -251, -252, einsteinium-253, -254, fermium-255, and so forth. And then he went on to this mendelevium experiment and finished his work with us in 1956. In the meantime, he has become one of the world's leading investigators in the chemistry, especially the ion exchange chemistry, of the actinide elements, having done some of the nicest work in the world in this field.

7.5.3.2. *Reminiscences: Dr. Choppin*

I am grateful to Prof. Seaborg for inviting me to participate in this anniversary symposium on mendelevium. Particularly helpful is the fact that he put my talk after that of Al Ghiorso and Bernie Harvey, who have recalled rather well the significant aspects of those days. Obviously, this relieves me of the obligation to say anything significant.

When I arrived at the Rad Lab in June 1953, it was with an incredible vacuum of knowledge about nuclear chemistry in general and heavy elements in particular. For the first few months I could not remember which was element 94 — americium or curium — since, obviously, the element after uranium, element 93, was plutonium. You perhaps can imagine how it was to be around Stan Thompson, Al, and Bernie as they threw around phrases like "citrate columns," "6.44 MeV," "15 barns," etc. That Glenn accepted me as a postdoctoral assistant with my background in liquid ammonia research is still a surprise, but I have always been grateful. Thank you, Glenn, very much.

So many things happened during the three years I worked with Stan, Al, and Bernie. It was long hours, with quick transitions

between many projects and fast trips to the MTR in Idaho. But it was even more a time of learning from three amazing teachers. A particular memory was one trip to the MTR to process the irradiated californium sample from the "Mike" shot debris. We were trying to "rediscover" element 100 so its formation could be published. Bernie and I opened the aluminum rabbit under the water in the reactor pit and discovered it was empty — no little quartz capsule with the californium and, hopefully, the fermium.

The day before, we had cleaned the trench of debris and, in doing so, had found a small quartz capsule, which we stored in a bucket in the trench in case anyone came looking for their lost experiment. Stan looked at this capsule and proclaimed it our californium sample! In fact, I doubt he had seen the californium capsule when Bernie and I prepared it three months earlier. Based on Stan's certainty, the capsule was returned to the reactor for several days' reactivation. Meanwhile the four of us went to Sun Valley to ski and to avoid being in touch with Glenn, who was more nervous about such things. We returned, betting on whether the capsule was, indeed, the californium sample or, maybe, cobalt — in which case we might spread ^{60}Co around. Of course, Stan's chemical intuition (or luck) held and we could report the formation of element 100.

In the experiments to make element 101, my primary responsibility involved the chemical separation for which we developed alpha-hydroxyisobutyrate as an eluant from columns of cation exchange resin. We needed something better than citrate or lactate to separate a few atoms of mendelevium from fermium. It seemed to me that α -But (our shorthand for α -hydroxyisobutyrate) might give us better separations. Stan agreed and called to campus, where an old bottle with about 2 g was found in the stockroom. Bernie and I, with the help of Docia McKenna and Marge Nervik, began studies which eventually led to performing several hundred column elutions over the next few months. For about the first month we had only those 2 g, so we had to recover and reuse the eluant solutions. Effects of α -But concentration, pH, temperature, column dimensions, loading

technique, flow rate, and drop size were among the variables we studied until we had a system which allowed us to separate a few atoms of mendelevium and to predict to the exact drop when those atoms would elute. We only had five fission counters to use for the peaks of the 101 and 100 elution as well as for the valley before, between and after the peaks. An additional problem was that einsteinium was the last actinide we could use to calibrate our elutions, and thus we had to rely on the rare earth analogs of fermium and mendelevium — erbium and thulium. Obviously, we solved our problems or we would not be celebrating this anniversary for another year or more.

One of my strongest memories of element 101 is not related directly to the discovery experiments, but rather to the repeat experiments that we did about six months later. At that time Otto Hahn had come to the United States on a visit as a result of an invitation by Glenn Seaborg and Ernest O. Lawrence. Hahn was giving a seminar in the chemistry department, and Glenn mentioned to him that element 101 was being rediscovered that night. He expressed an interest in watching some of the experiment, so we were told he would probably come around for a few minutes. Hahn came, watched, and poked into everything and stayed and stayed. Lawrence and Seaborg were getting a little tired and suggested leaving, but Hahn said he wished to watch some of the decays. He was like a five-year-old kid in his curiosity, and it was very impressive to see such an eminent scientist still excited about an experiment. This was quite an experience for me, because there I was in this very small counting room which was half-molar in Nobel Prize winners.

In conclusion, this is an opportunity to express my great personal debt to Stan, Al, Bernie, and Glenn, from whom I learned so much. Their consideration and friendship are the best memories of those days.

Glenn would come to the lab each afternoon and casually ask how things were going. I soon learned that he understood all and forgot nothing. The luncheon discussions in his office taught me much besides heavy element chemistry.

From Stan Thompson I learned that intuition beats careful thinking much of the time. Stan must have been one of the world's greatest instinctive chemists. Time and again the unexpected would happen and Stan would pull something out of his memory to suggest we try. Of course, as a new Ph.D. I knew "Chemistry" and I would often explain why that would not work. But we would try it and it did work. Stan taught me not to rely too much on books, but instead to go ahead and try an unlikely technique or experiment. Working with Stan Thompson was one of the finest things that have happened to me.

I learned from Al that enthusiasm and energy are absolute prerequisites if you are going to get very far in research. I was constantly amazed that he never ran out of ideas, he never ran out of energy, and he was always arguing about everything. Along the way he taught me a great deal about politics. After cyclotron runs sometimes Al and Bernie would drive me home. Often we would stop, about 2–3 a.m., for coffee and Al would lecture me that I had to be more liberal in my thinking. I have tried to keep this as well as the other lessons that Al taught me as to how to go about doing science while enjoying it thoroughly.

But if I had come to believe that instinct and enthusiasm were enough, it would have been an inadequate education, so I have always been grateful that Bernie was part of the group. First, although he was quite experienced he had just come to Berkeley. So, as the new members of the group, we developed a close relationship — possibly as a slight defense against the two old pros — Stan and Al. Bernie combined careful, logical thinking and deep understanding. The same precision, care, and understanding were evident in his laboratory work.

So each of my coworkers affected me greatly and I thank them for that. I also thank them for the three exciting years during which mendelevium was born.

7.5.3.3. Comments: Dr. Seaborg

I'm glad you brought up this capacity of Stan Thompson for intuitive thinking in chemistry. He had the best — and I hesitate to say this in German, because Mike Nitschke is here — *chemisches gefühl* of anybody that I know. That was true throughout his career, especially at Chicago, at the Metallurgical Laboratory, when we developed the chemical process for the isolation of plutonium. This was of extraordinary importance for the free world at that time. He was the moving spirit behind that process, the originator of the process which wasn't supposed to work. Our main problem at Chicago was that we were working on something that was considered to be impossible. It just didn't follow the principles of chemistry, but Stan kept saying, This'll go, it's OK. And it was probably the only process that we could have put through in the time at our disposal.

Also, your mention of Otto Hahn reminds me of another story. In 1966, when he must have been 87 or 88 years old, he came to Vienna to receive the Fermi Prize, and I had the responsibility and pleasure as Chairman of the Atomic Energy Commission to bestow this upon him in a huge ceremony. After the ceremony and later in the evening, when most of us were getting kind of tired and wanted to retire, he wanted to go out to the bar. And then, believe me, he did have an eye for the girls. And I think we finally had to say, you know, Prof. Hahn, don't you think maybe it is getting a little late for a man of your age. I guess he finally humored us and called it quits.

I should mention that Greg Choppin traveled the farthest of anybody in coming to this meeting. He is at Karlsruhe on sabbatical leave this year at the Institute for Transuranium Research, so we appreciate very much his taking the trouble to come this distance.

7.6. Conclusion

After hearing from the discoverers, the symposium continued with the papers by Hulet, Hoffman, and Friedman on the current state of the knowledge of mendelevium. A fitting close to this chapter is

Greg Choppin's account of "The Fire Bell Incident," which he included in the final chapter of a 1978 high school chemistry textbook⁷⁵ which he coauthored.

"The night of the actual discovery experiment, not only were the four of us — Ghiorso, Thompson, Harvey, and myself — present, but also perhaps a half dozen other people, including the operators of the cyclotron, the technicians helping with the experiment, and people concerned with the radiation safety aspects. Our late hours had led us to set up a small cooking arrangement in one of the laboratories to prepare bacon and eggs before going home each morning. Since we were quite anxious to know if we had succeeded, we were reluctant to have the radioactive decay events occur without our knowledge while we were eating breakfast.

"Ghiorso solved the problem by connecting the radiation counter to the fire alarm bell for the laboratory. Accordingly, soon after we started breakfast, the fire alarm bell went off. We all gave a loud enthusiastic cheer, since it meant that we had succeeded in synthesizing element 101. Ghiorso and Harvey raced down to the counting room to turn off the fire alarm bell. They also expressed their excitement by writing on the recorder chart connected to the counter. Bernie Harvey wrote 'Hooray' on the chart beside the deflection, the first comment on the synthesis of element 101. When the fire alarm bell went off a second time, indicating that a second atom of 101 had decayed, Bernie wrote 'Double Hooray,' and after the next deflection, he wrote 'Triple Hooray.' The synthesis continued and other decays also occurred. But after the Triple Hooray, we knew that element 101 was definite, and we became somewhat more restrained in our enthusiasm. I was cleaning the laboratory from the experiments when Ghiorso and Harvey signed their names to the statement that this conclusively proved the identification of 101.

"About 4:00 a.m., we all went home in a state of enthusiasm and excitement. I got very little sleep that morning. That isn't surprising, since participating in the discovery of a new element, which you know will be recorded in chemistry books as long as there are

chemistry books, is a rather intoxicating experience for a 27-year-old postgraduate student just beginning his scientific career.

"We had agreed to reassemble at the lab shortly after lunchtime the next day to begin to write a publication that we would send to the scientific journal *Physical Review* to inform the scientific world of our discovery. However, about 10:00 a.m., an event occurred while we were home sleeping that is still legend at the Radiation Laboratory. Ghiorso had unintentionally left the counter hooked to the fire alarm bell. A single atom, which should have decayed much sooner, fissioned that morning. It triggered the fire alarm bell, and since several thousand people at the Radiation Lab did not realize that the bell was hooked to the counter, they assumed that a fire had broken out. The laboratory operates with a volunteer fire force, and everyone in the lab is quite concerned about any fire, since there is so much radioactive material around. The result was a wild milling of people running up and down the hallways of the chemistry building looking for the fire. This state of affairs lasted for some 15 or 20 minutes, until somebody traced the wire for the fire bell to the counter and learned the source of the 'fire.' The result of this incident was that E.O. Lawrence, the director of the laboratory and the man who had won a Nobel Prize for the invention of the cyclotron, sent Glenn Seaborg, as the director of the chemistry division, an official note of congratulations on the discovery of element 101. He appended a statement saying it was official laboratory policy that even for so important an experiment as the discovery of a new element, fire alarm bells were not to be hooked to detectors.

"During the many discussions when we used to sit around and talk about everything under the sun, including the experiments with which we were involved, we had decided that it would be appropriate to name the new element, which marked the beginning of the second hundred elements of the periodic table, after the man most responsible for the periodic table, Dimitri Mendeleev. That name was accepted by the International Union of Pure and Applied Chemistry (IUPAC), which made it official."

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Chapter 8

Nobelium and Lawrencium

8.1. Introduction

After the discovery of mendelevium it was recognized that elements of higher Z would have to be made by ions with Z greater than that of ^4He since there were no suitable targets beyond element 99. At that time there were only three accelerators in the world that were capable of making such bombardments: the new HILAC at Berkeley which was just coming on line, the cyclotron at the Kurchatov Institute in Moscow, and the cyclotron at the Nobel Institute for Physics in Stockholm. The Stockholm laboratory, the first to try to surmount this barrier, unfortunately made a grievous error and it appears now that the activity that they claimed to have identified in 1957^{8.1} as coming from element 102 was only an artifact of their experiment. Furthermore, experiments conducted at Berkeley in 1968^{8.2} showed that the 3+ oxidation state of element 102 is not the most stable one in aqueous solution as the Stockholm group had assumed, and there was no way that it could have been chemically isolated in their experiments. Their unfortunate mistake set the tone for the subsequent initial discoveries by the other contenders, both American and Russian; the needed technology had not yet matured and it took a number of years for the groups at Berkeley and Dubna to correctly sort out the rather confusing complex of nobelium and lawrencium isotopes. This chapter tells the story of how that was done.

8.2. Nobelium (Element 102)

8.2.1. The Nobel Institute Experiment of 1957

In 1957, possibly emboldened by the Berkeley *tour de force* two years before in which element 101, mendelevium, was discovered, in the first “one atom at a time” experiments, Paul Fields and Arnold Friedman, nuclear chemists at the Argonne National Laboratory, brought together scientists from the Atomic Energy Research Establishment at Harwell and the Nobel Institute for Physics in Stockholm to try to discover element 102. Since Argonne had no suitable accelerator of its own at that time, their plan was to export curium from the U.S. to the Harwell Laboratory, where suitable targets would be made by painting the material from a solution of the nitrate in a mixture of acetone and amyl acetate onto thin aluminum foil in a 0.5-cm² area. These targets would then be taken to Stockholm and bombarded with an internal beam of 90–100 MeV ¹³C ions using a special probe at the Nobel Institute Cyclotron. Eventually, they produced some six targets with the isotopic composition, 95%–244 and 5%–246, each with a surface density of about 1 mg/cm². The ²⁴⁴Cm isotope is a highly radioactive α -emitter having a half-life of only 20 years, so this would not be an easy experiment!

The Fields team first tried short bombardments, followed by the very simple nonchemical experimental procedure of catching recoils from the target in a plastic foil and then analyzing it for α -activity. After a bombardment of about 30 min, the catcher foil was placed on a platinum plate and ignited to remove the bulk of the organic material. This plate was then placed inside a gridded α -particle ionization chamber for energy analysis — and there they observed an occasional α -count with an energy of about 8.5 MeV! The amount of activity was minuscule, however, and could only be measured in terms of isolated counts. The results of the bombardments were erratic, only three of the six targets producing the 8.5-MeV α -activity. This problem was blamed on recoil absorption caused by deterioration

of the targets from α -radiation and the ^{13}C beam. From the profile of the very few events that they did observe they decided that the 8.5-MeV α -emitter had a half-life of roughly 10 min.

They next decided to use a chemical procedure to identify the atomic number of the "activity" that they had found, since the half-life seemed to be long enough for this purpose. Their procedure consisted of dissolving the activity from the platinum plates and sorbing it on a cation exchange resin column, since they expected that element 102 would be in the 3+ state and thus, like the other heavy actinides, would be eluted from the column with an ammonium α -hydroxyisobutyrate solution before those with lower atomic number. [They were unaware that it had been predicted that a 2+ oxidation state might exist for element 102 by analogy to its rare earth homolog, ytterbium, which can be reduced to 2+ in aqueous solution, and their chemistry was not designed to allow for this possibility.] In these experiments they observed a few events that they attributed to their 10-min, 8.5-MeV α -activity at the "expected" elution position for element 102. They also reported that the 8.5-MeV α -activity eluted from a hot cation exchange resin column with 6 M HCl together with californium and fermium activities produced in these irradiations. They concluded that the 8.5-MeV α -activity was due to an isotope of element 102 with mass number most likely either 251 or 253, either formed directly, or as a result of *ec* decay of an element 102 isotope with a 10-min half-life to a much shorter-lived α -emitting mendelevium daughter. This done to their satisfaction, they sent off their findings to *The Physical Review* as a Letter to the Editor.^{8,1} In the letter they suggested "the name nobelium, symbol No, for the new element in recognition of Alfred Nobel's support of scientific research and after the institute where the work was done." Some months later the name was accepted by the IUPAC. If true, it would have been a spectacular piece of work. It was not true and in the end all that remained was the name!

Just by chance, Ghiorso happened to be in Seaborg's office at the moment when the news first came to Berkeley by telephone from Sweden that element 102 had been found. Ghiorso was puzzled by

the unexpectedly high α -energy and the long half-life that had been observed, and immediately wondered whether the Fields team could have been confused by emissions from known elements. He could see from the Chart of the Nuclides that hung in Glenn's office that a possible candidate was 8.5-min ^{225}Th . He and Wayne Meinke, a Seaborg graduate student, had recently discovered this isotope in bombardments of ^{232}Th with high energy helium ions at the 184-Inch Cyclotron to produce 58-min ^{229}U which α -decayed to ^{225}Th . They had found that, after a succession of three very short-lived α -decays, the chain reaches 4- μs ^{213}Po , whose α -energy of 8.38 MeV is not very far from the putative 8.5 MeV observed by the Nobel Institute team. The immediate question was whether that isotope could be formed by 100-MeV ^{13}C bombardment of curium. Strong doubts that the element 102 experiment was correct were aroused immediately by this possibility, so it became urgent to see if the proposed reaction could take place.



Fig. 8.1. HILAC Post-Stripper tank on its way up the hill to the Radiation Laboratory, 1955.

The Berkeley HILAC, having just been completed in October 1957 (Fig. 8.1), was undergoing commissioning tests, but the nuclear chemists had not yet performed any experiments with it. That very evening Ghiorso and Stan Thompson decided to see if they could produce ^{225}Th by bombarding a thick foil of uranium metal with the 10-MeV/A ^{14}N ion beam that the accelerator people had been using for test purposes. A simple chemical procedure to get rid of the uranium was all that was needed and after an hour's bombardment with the 2-pulse/s ^{14}N beam, an α -pulse analysis of the actinide element fraction quickly showed that they had, indeed, formed a large number of isotopes in this region including the hypothesized ^{225}Th ! Although this was not proof positive, it was an indication that some of these nuclides might have been formed also in the Swedish experiment.

They then conducted experiments with a curium target over a period of some four months to look for the 10-min, 8.5-MeV α -activity. They had important advantages over the Stockholm team that gave them orders of magnitude more sensitivity. First of all, the Berkeley HILAC was able to deliver intense external beams of monoenergetic heavy ions. On the other hand, at the Nobel Institute the experimenters had to cope with weak beams with a wide energy spread *inside* the cyclotron. These restrictions severely limited the kinds of experiments that could be conducted at the Nobel Institute Cyclotron in 1957.

When the Berkeley team of Ghiorso, Torbjorn Sikkeland, John Walton, and Seaborg set out to try and confirm these findings, it was assumed that they were correct since the Stockholm team had published its work in a well-written paper in the peer-reviewed, highly respected journal *The Physical Review*. Ghiorso and his colleagues felt that their most straightforward approach was to use a thin palladium catcher foil to catch the recoils that resulted from the bombardment of ^{244}Cm by ^{13}C and, after its dissolution, to pass the solution through an anion column to remove the palladium. The resultant trivalent actinide fraction which ran through the column could then be analyzed for its α -activity. This procedure they were

able to do repeatedly in about five minutes. The result was that, even though they saw, very abundantly, the well-known actinide nuclides, ^{245}Cf , ^{246}Cf , and ^{250}Fm , in amounts that were a hundred times more intense than those observed in the cyclotron experiments, they never observed even a single 10-min, 8.5-MeV α . These experiments chemically duplicated those of the Stockholm group but with far higher sensitivity and showed that no 3+ actinide was produced with an α -energy of 8.5 MeV. They thus fulfilled their objective but, *ironically, like the Stockholm experiments, their procedure would not have separated out element 102 either, since the later Berkeley work^{8.2} showed that element 102 would have been in its most stable 2+ state for those experiments!*

The Berkeley experimenters asked themselves if they could be doing something wrong. Maybe the chemistry of this new element was unusual. Could it be that metallic atoms of element 102 were extremely volatile and were vaporizing out of the palladium foil, made warm by beam heating? To address this possibility they resorted to a second method, catching the recoils in a thin mylar foil cooled by helium gas. They reasoned that the element 102 recoils would immediately oxidize inside the plastic foil and thus not be volatile. After bombardment, the mylar catcher foil was placed on a platinum plate, where it was ignited to burn off the plastic, just as was done in Sweden, and then analyzed. Again large amounts of the known actinides were seen but no 8.5-MeV alphas.

A third method was tried, which they were confident would not discriminate in any way against the ephemeral activity. A negatively charged plate was mounted in the target chamber in an atmospheric helium environment to attract the positively charged recoil ions that stopped in the helium during the bombardment. Afterward, this plate was analyzed in an external gridded α -chamber and, again, they saw large quantities of the actinides but no α -particles with an energy of 8.5 MeV.

A couple of years later a variant of this method was used in which the recoils were stopped in helium gas and collected electrostatically on a conducting tape. The tape periodically transported the

collected ions to a special large on-line grid chamber that had been developed for this purpose. As will be shown later, this device was successful in discovering 2.3-s ^{252}No , but no relatively long-lived 8.5 MeV alphas ever showed up. All of these experiments were without success of any kind in detecting the Stockholm "ghost" even though the Berkeley sensitivity exceeded that of the Fields team by two orders of magnitude. Some years later, to give the *coup de grace* to the whole idea, it was shown^{8,2} that *the 2+ state of element 102 is the most stable species* in aqueous solution and thus could not possibly have eluted early from a cation exchange resin column under the chemical conditions used.

The question naturally arises as to how such a gross mistake could have gone uncorrected. In 1959, when Ghiorso looked at the original data in England with John Milsted, the Harwell Laboratory member of the team, Ghiorso reported being appalled by how poor it was. The amount of activity at all energies being observed in any one of the individual experiments was never more than a few events. Most of the time no 8.5-MeV events were recorded, with only 12 out of the 50 experiments being "successful"; their will to believe was very strong, however, and the team became blinded to the possibility that their work was completely wrong. It is unfortunate that they did not subject their work to some sort of review by knowledgeable scientists in the field of heavy elements. This could have been done without jeopardizing their priority of discovery.

After the Berkeley group had gone ahead and found actual element 102 α - and SF-activities as outlined later, it was suggested that the Stockholm group should withdraw the name "nobelium." This they refused to do, hoping that somehow Berkeley had not used the proper conditions to make their 10-min, 8.5-MeV activity and that eventually their work would prove to be correct. Some ten years later, because the euphonious name "nobelium" had come into general usage, Ghiorso and Torbjorn Sikkeland, in an article in *Physics Today*,^{8,3} relented and suggested that the name "nobelium" remain. For a long time, though, they used to say that whenever they saw the symbol No they thought of *nobelievium* rather than nobelium!

However, this was not the only controversy that would arise over this element. A new one would gradually develop between Berkeley and Dubna, the difficulties in this case being caused by the fact that two isotopes of element 102 were involved in the discoveries by the Berkeley team and a third by Dubna and this was not realized until better techniques gradually evolved that could answer the questions that were raised by the research. It is important to remember that the methods used for nuclear identification at this time were still being developed so that it was not unusual for mistakes of interpretation to be made by all groups working in the field.

In this process of unraveling the mysteries during the period 1963–1966, various Dubna groups under the general direction of Prof. G.N. Flerov added valuable information to the knowledge of the element so that the combined work of Berkeley and Dubna resulted in a complete clarification of the isotopes of element 102 from mass number 251 to 259. It is for this reason that Berkeley recognizes Dubna as a codiscoverer of element 102. The following account discusses in detail the various events that followed the initial fiasco at Stockholm.

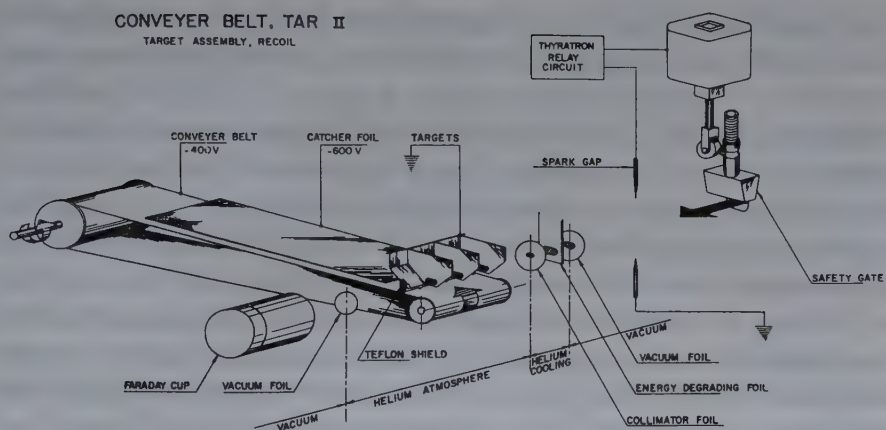
8.2.2. The Berkeley Work

After Berkeley had become certain that the Nobel Institute work was completely wrong they turned their focus to experiments that might be able to find an actual isotope of element 102. From the experiments that they had already performed in looking for the nonexistent activity, they knew that it was not going to be easy and did not think that at this time they could take advantage of chemistry and find rare atoms of the new element directly as was done in the mendelevium experiment. Instead, they turned their attention to physical methods. Experiments with lighter elements and calculations seemed to indicate that the most likely element 102 product of the reaction of ^{12}C ions with a ^{246}Cm target would be from the 4n-out reaction to make $^{254}102$. This prediction was not completely correct, as it turned out, but it seemed reasonable at the time.

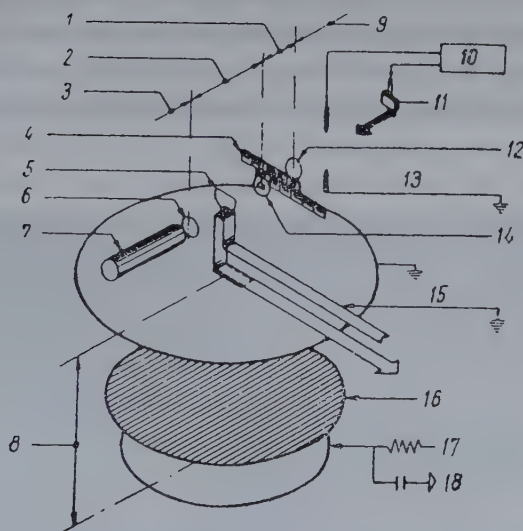
Accordingly, they concentrated their attention on the relatively long-lived, α -emitting daughter of $^{254}\text{102}$, ^{250}Fm , since its 30-min half-life was long enough to allow chemical identification. If they could devise some method of connecting the element 102 mother with its known element 100 daughter, they could make a positive identification.

The 184-Inch Cyclotron experiments of Meinke and Ghiorso had shown that recoil after α -decay could be a powerful tool for this purpose just as it was in the early days of the research with natural radioactivities, so it was decided to try it out. The idea was that a negatively charged plate could be used to collect beam-produced transmutation recoils that had been stopped in an atmosphere of helium as positive ions; once these atoms were on the collector plate, it should be possible to collect those daughter atoms, which had recoiled after α -decay of the mother element 102, onto another plate which was biased even more negatively. These atoms would be dissolved and ordinary chemical methods could then be used to identify the atomic number of the daughter and hence the mother. This would be a first step; Ghiorso had in mind a more complicated apparatus for detecting the mother directly but he knew that it would take some time to develop the apparatus. The "milking machine" would have to come first.

A double-recoil geometry was devised to accomplish this seemingly magical feat. A metallic conveyor belt with a negative bias was placed in an atmosphere of helium just below three sequential targets of curium to collect the primary recoils of element 102 that were ejected by the reaction of ^{12}C ions with the target. A more negatively biased collector was set up just above the belt and downstream from the target. This would collect the positively ionized element 100 daughter atoms that were kicked off the belt by recoil from the α -decay of any element 102 atoms that emitted their α -particles into the belt. After a suitable bombardment, this collector would then be sectioned into five pieces to be analyzed. In the chemical experiments necessary for identifying the atomic number of the daughter, the activity would be dissolved from the collectors.



(a)



(b)

Fig. 8.2. Alpha recoil system. (a) Schematic diagram of the conveyor belt experiment used in the original Berkeley research on element 102. Two isotopes of that element, with masses 254 and 252, complicated the analysis and temporarily led the researchers astray. (b) The Berkeley on-line grid chamber which was used for the discovery of the α and SF emissions from $^{252}_{102}$. Unfortunately, its life was cut short by a disastrous target breakage incident.

This was the way that the equipment (see Fig. 8.2) was designed to work in principle; how would it perform? The equipment was tested first with a stand-in reaction using a target of ^{240}Pu to make an unknown isotope, ^{248}Fm . Ghiorso and Sikkeland were very excited when, as expected, its known daughter, 20-min ^{244}Cf , was found to be transferred to the catcher foils with a half-life of 0.6 min. This experiment inaugurated a new era of identifying isotopes in the transuranium field by their genetic relationships.

Having demonstrated that the device worked, the group repeated the experiment with curium as a target having the same isotopic composition (95% ^{244}Cm ; 4.5% ^{246}Cm) that was used in the ill-fated Stockholm experiments. The cross section for the $^{246}\text{Cm}(^{12}\text{C},4n)^{254}102$ reaction was relatively low and this made the experiments difficult in those days before the advent of computers. Since it was necessary to set up a system that would analyze the five pieces of catcher foil at once, a system of five gridded-alpha chambers (Fig. 8.3) and their associated amplifiers was assembled. To collect the information as it

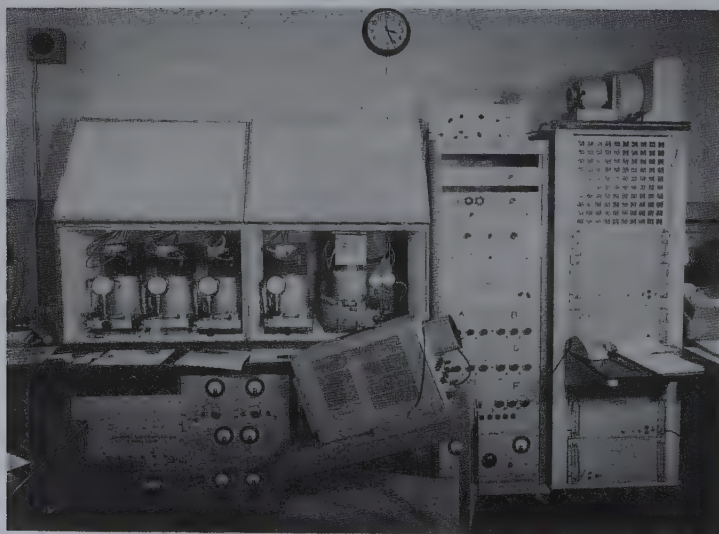


Fig. 8.3. System of five gridded alpha chambers.

occurred, a paper printer system that would record each event as it was produced in terms of time, pulse amplitude, and origin was used.

It was a slow and clumsy system but, at the low counting rates that they had to contend with, it was workable. Problems of stability and resolution, however, were a big headache, aggravated because there were five units going at once. A series of runs showed that α -particles with energies approximately that of ^{250}Fm did appear on the catcher foils with a time distribution indicating a half-life of about 3 s. An excitation function showed that the most activity was transferred when the ^{12}C beam energy was 70 ± 5 MeV. This value was close to that calculated by Sikkeland using a method that he had developed a little earlier for this purpose. That these atoms were actually transferred by α -recoil was proven by the fact that ^{246}Cf was not observed on the catchers in spite of the fact that it was found in great abundance on the conveyor belt. When the belt speed was changed, the time distribution changed accordingly. All of this information was obtained by pulse-analyzing the catchers directly.

In separate experiments, the activity on the catcher foils was removed, sorbed on a cation exchange resin column and eluted with α -hydroxyisobutyrate. As expected, ^{250}Fm was found in the element 100 fraction. Since all of the catchers were combined for the chemical experiments, no half-life information was derived in this case. At this point, the work was published in *Physical Review Letters*^{8.4} in July 1958 by Ghiorso, Sikkeland, John R. Walton, and Seaborg. At the same time, a separate Letter was published by the same authors showing that, even though they had orders of magnitude more sensitivity, the 1957 work on element 102 at the Nobel Institute could not be confirmed.^{8.5}

It was assumed that the half-life of $^{254}102$ must be 3 s, since that was the decay period observed without chemistry. In actuality, it has a half-life of 55 s, as would be shown by a Dubna group several years later, so it would have distributed its ^{250}Fm daughter across all five of the collectors. What the Berkeley group did not know at that time was that a different element 102 isotope, $^{252}102$, with a

half-life of 2.3 s, was also present on the belt, having been produced by a ($^{12}\text{C},4n$) reaction with the 95%-abundant ^{244}Cm isotope in the target. The $^{252}_{102}$ that was produced in this way would transfer its 36-s daughter, ^{248}Fm , which then would α -decay to 19-min ^{244}Cf ($E_{\alpha} = 7.21$ MeV). It would be this activity that they had mistaken for the 30-min ^{250}Fm (7.43 MeV) in the milking experiments made without chemistry.

Why was this mistake made? It is hard to know exactly why, but a good guess is that the Berkeley team was blinded by its success in showing chemically that they had milked ^{250}Fm by α -recoil. Thus when they analyzed the activity on the collector without chemistry they were prepared to believe that it was just more ^{250}Fm . The two descendants, ^{250}Fm and ^{244}Cf , have similar half-lives and α -energies so that it was possible to confuse the two, using the instruments of the time, *especially when only small amounts of activity were involved*. It is known now that *the fermium and the californium isotopes had to be present at the same time* and that since the ^{244}Cf had an α -energy only 200 keV lower in energy it would appear on the tail of the ^{250}Fm - α -energy spectrum. It could easily have been included as part of the fermium spectrum, especially when a wide energy region was included in the summation of the activity that had been transferred.

After the milking experiments had been completed, the next step was to construct a large on-line Frisch grid chamber that would permit measurement of the energy of this 3-s α -emitter. (There simply was no other way of making such a measurement, since this was some two years before the development of solid-state alpha detectors). It was found by careful experimentation that an Ar/10% CH_4 mixture at atmospheric pressure would work satisfactorily (because of the difference in gas densities) with the chamber in an upside-down position a few centimeters below the helium-filled target chamber. An aluminized mylar tape passing through slits acted as a conveyor belt to carry the recoils the short distance from where they were collected in helium gas at atmospheric pressure to the analyzing position in the Ar/ CH_4 -filled grid chamber below.

The first test with this equipment was with the ^{240}Pu target to make the same 36-s ^{248}Fm that had been discovered genetically in the first milking experiment tests. They observed its α -particles this time and measured an energy of 7.8 MeV. In later work with this isotope using silicon crystal detectors, it was found that their true energy was 7.9 MeV. This provided an internal calibration for these early grid chamber experiments and indicated that the subsequent measurement for $^{252}102$ must be increased by 0.1 MeV, from 8.3 to 8.4 MeV.

The next step was to try curium as a target. The Berkeley group immediately ran into what was to become the ubiquitous problem of lead contamination that would plague all such experiments that tried to measure α -radiation from the transuranium elements. The difficulty stems from the fact that heavy ion bombardment of even nanogram amounts of lead produces copious quantities of α -emission from isotopes of the elements just above lead in the nuclide chart. Because these emissions overlap the heavy element α -emitters in energy and half-life, this contamination often proved to be a very serious obstacle. It took a lot of hard work by chemists Bob Latimer and Jim Harris to purify the curium sufficiently so that it could be used for this research.

As soon as the bombardments had begun, the group observed a new α -particle activity with an energy of 8.3 MeV and a half-life of about 3 s. This was very exhilarating to them — because now all of their observations seemed to fit together to form a consistent picture! They immediately drew what seemed to be the obvious conclusion, that this must be the same α -activity which had given rise to the ^{250}Fm that they had observed in the previous milking experiments and, therefore, must be $^{254}102$. Along with the α -particles, they observed a very prominent ($\approx 30\%$) SF activity with the same 3-s half-life. Encountering intense SF activity was a major surprise, so they went to a lot of trouble, first to make sure that they were really SFs, and then that they came from the same isotope that emitted the α -particles. No matter what they did the ratio of SF/ α could not be

changed, so it was finally concluded that this must be the case. Thus it was duly reported^{8,6} by the Berkeley group in 1959 that $^{254}\text{102}$ was a 3-s, 8.3-MeV α -emitter with a very prominent SF branch of 30%.

That *two* isotopes of element 102, with mass numbers 254 and 252, were actually produced in these carbon bombardments was not realized until several years later when a Dubna group showed that the half-life of $^{254}\text{102}$ was 55 s not 3 s, and did not have an easily observable SF branch! Without question, further runs with the grid chamber normally would have uncovered the fact that two isotopes of element 102 were involved. Unfortunately, in 1959 there was a disastrous accident with the apparatus that destroyed all of the precious curium targets and contaminated the grid chamber so heavily that it was unusable. Lack of confidence in their ability to handle highly radioactive targets kept the Berkeley group "gun-shy" for a considerable time afterwards, so there was no urgent desire to repeat the curium experiments immediately.

Since the nobelium research seemed to be internally consistent and without any obvious flaws, the group decided to press on and look for element 103. They felt that they had a good chance to produce that element by using boron ions to bombard the few micrograms of californium that by now had been produced in the Idaho Materials Testing Reactor facility. In 1961, as a part of the identification of the 8.6-MeV α -emitter of element 103 using silicon crystal α -detectors, it was necessary to carry out an extensive set of cross bombardments with other targets and projectiles. During some of these bombardments, they observed an activity with a complex α -spectrum around 8.2 MeV with a half-life of about 15 s and deduced that it could only be due to element 102. Because the californium target had four isotopes, they were unable to pin down the mass assignment and temporarily assigned it to $^{255}\text{102}$. It later turned out to be due to $^{257}\text{102}$ with, more accurately, a half-life of 26 s and α -groups at 8.32, 8.27, and 8.22 MeV.

8.2.3. The Dubna Work

This was the situation up to 1961. There had been sporadic attempts as early as 1957 by G.N. Flerov and his colleagues^{8,7} to produce element 102 by bombardments of ^{239}Pu and ^{241}Pu with ^{16}O ions at an old cyclotron at the Kurchatov Institute in Moscow. The attempts had poor selectivity because of their use of nuclear emulsions for α -particle range measurements and were hindered by strong backgrounds caused by lead impurities; there was no way to differentiate element 102 from the extensive background. The work was then transferred to the recently completed Dubna Laboratory, where all subsequent heavy ion experiments would be carried out.

The Dubna Laboratory was intended to be the joint central nuclear research establishment for the use of the Soviet Bloc countries, the equivalent of CERN, the joint central laboratory for the Central European Community. The Dubna Joint Institute for Nuclear Research is situated on the Volga River in a beautiful birch forest some 100 km north of Moscow. At this site, two heavy ion cyclotrons were constructed, a small sector-focused machine with a 1.5-m pole tip diameter, and another, very large conventional machine with 3-m pole tips. For the time, they were both excellent accelerators; in particular, the large one was capable of accelerating very large quantities of beam internally because it could use a very low charge state from the ion source to reach the energies needed. Efficient extraction of the beams was not possible from this machine, so to make the maximum use of the beams the Dubna scientists usually placed their experiments inside the cyclotrons. This both severely limited what kinds of experiments could be done and increased the difficulty of performing them.

In 1964, in the first reliable work on element 102 at Dubna, E.D. Donetz, Shchegolev, and Ermakov^{8,8} bombarded ^{238}U with ^{22}Ne ions and discovered ^{256}No by chemically identifying its 23-h recoil daughter, ^{252}Fm . This isotope was separated from its parent after α -particle decay by the use of a version of the Berkeley double recoil principle, using a rotating wheel rather than a belt. It was a well-

done experiment in which they chemically separated the fermium daughter from the α -recoil fraction and seemed to have good statistics; the strange thing about this work was that they obtained a half-life for the mother ^{256}No that was around 8 s!

In 1967, V.A. Druin *et al.*,^{8,9} in bombardments of ^{nat}U with ^{22}Ne , used a helium jet system to transport activities to a catcher foil which was shuttled to a solid state detector; they found an 8.41-MeV α -group with a half-life of 6 ± 2 s, which they attributed to ^{256}No . In 1968, Flerov *et al.*^{8,10} published a paper on the results from bombardments of ^{242}Pu by ^{18}O ions as well as the previous reaction, and reported an improved half-life value of 3.7 ± 0.5 s for the 8.42-MeV α -group. More than two decades later, the Hoffman group^{8,11} at Berkeley made very accurate measurements of the half-life of ^{256}No , produced via the $^{248}\text{Cm}(^{12}\text{C}, 4n)$ reaction, and remeasured the energy of the principal α -group as 8.448 ± 0.006 MeV. They found its half-life to be 2.91 ± 0.05 s. They confirmed that they were observing ^{256}No by using the α -recoil technique to identify its ^{252}Fm daughter. Based on the consistency of their measured SF half-life of 3.0 ± 0.2 s with the α -half-life, and the relative constancy of the SF/ α ratio of 0.0053 ± 0.0006 from different experiments at somewhat different energies, they also assigned the observed SF activity to ^{256}No . The new values of the half-life and the α -decay energy, although more precise, were in essential agreement with the 1967 measurements^{8,12} of Ghiorso, Sikkeland, and Nurmia and the 1968 measurement by the Dubna group of 3.7 ± 0.5 s for the decay of the 8.43-MeV α -group.

In 1965 Donetz *et al.*^{8,13} used the same double recoil apparatus to determine the half-life of ^{254}No , and much to the chagrin of the Berkeley experimenters, found that it was 50 s, not 3 s as claimed by Berkeley. Again the experiment was well done, the milking of the 30-min ^{250}Fm being accomplished with dispatch and efficiency. In this same year, B.A. Zager *et al.*^{8,14} made a measurement of ^{254}No using silicon detectors for the first time at Dubna and found that the half-life was between 20 and 50 s and that the α -energy was 8.10 MeV. In 1967, V.L. Mikheev *et al.*^{8,15} took giant strides forward

by confirming the α -energy for ^{254}No and finding α -activities that they assigned to 95-s ^{253}No and ^{252}No . The latter isotope had an α -energy of 8.41 MeV and a half-life of 4.5 ± 1.5 s, with no SF branching reported. Later in the same year, Druin *et al.*^{8,16} discovered another new isotope of element 102, the 3-min ^{255}No . This isotope was very important, because its half-life is long enough to permit studies of the chemical properties of nobelium, such as the determination of its most stable oxidation state in aqueous solution.

8.2.4. The New Berkeley Work

The Berkeley group, dismayed by these new results, realized that the 1959 accident had been even more disastrous than they had thought, and decided that they had to find out immediately whether Dubna was right in *every* respect. This was in 1966, at a time when they were in the midst of developing an innovative wheel/gas jet system equipped with four silicon detectors intended for use in a search for element 104. This development was interrupted and, in an intense series of bombardments, they bombarded essentially monoisotopic targets of curium with mass numbers 244, 246, and 248 with carbon ions of mass numbers 12 and 13 to produce all of the accessible element 102 isotopes. In a very short time, with the new system and the large cross sections of $0.1\text{--}1\ \mu\text{b}$ for these reactions, they were able to reidentify accurately the nobelium isotopes with mass numbers 251–258 to settle to their satisfaction the uncertainties that had developed between Dubna and Berkeley in energies, half-lives, and mass number assignments. With the exception of ^{256}No , where they found its half-life to be 3 s rather than the 8 or 9 s originally found by Dubna, there was reasonable agreement between the two laboratories. The Berkeley group was mortified to find that the most important prior discrepancy, the half-life of ^{254}No , was cleared up when they found that the Dubna result was quite correct; the half-life was 55 s, not 3 s, and it did not undergo prominent SF decay. *It was ^{252}No that had the 3-s half-life and underwent SF 30% of the time.* It was at this point that they realized that they had been fooled in

1958–1959 by the simultaneous presence of two isotopes of nobelium, ^{254}No and ^{252}No .

The element 102 early work is obviously quite complicated, so to clarify what happened it has been summarized chronologically as follows:

(a) The Berkeley group showed^{8,5} within a few months of its publication that the 1957 work at the Nobel Institute was completely in error. The 10-min, 8.5-MeV α -activity could not be produced by any heavy-ion reaction on curium.

(b) The 1957 work^{8,7} at the Kurchatov Institute was inconclusive at best because of resolution and background problems and incompatible with presently known data on nobelium isotopes.

(c) In 1958 at Berkeley, the isotope ^{250}Fm was shown by chemical means to exist in an α -recoil-milked fraction,^{8,4} thus establishing the presence of $^{254}102$.

(d) In 1959 at Berkeley, the isotope $^{252}102$ was discovered as an ≈ 3 -s α -emitter with a strong SF branch.^{8,6} The linkage of E_α , SF, and $T_{1/2}$ for this activity uniquely identified it as the belonging to the isotope $^{252}102$.

(e) In 1961 at Berkeley, the isotope $^{257}102$ was discovered as a 15-s α -emitter during the element 103 research.^{8,17} It was arbitrarily assigned the mass number 255 at the time; it is now known to be 257, a better half-life for it being 25 s, as shown by Pirkko Eskola *et al.*^{8,18}

(f) In 1964 at Dubna, the isotope $^{256}102$ was discovered and identified (with a reported half-life of 9 s, now known to be 2.9 s) by the recoil/chemical milking^{8,19} of its fermium daughter, ^{252}Fm . In 1996, the same group discovered^{8,20} that $^{254}102$ has a 55 s half-life, not 3 s as Berkeley had claimed. The Dubna group also discovered^{8,21} 3-min $^{255}102$ and 105-s $^{253}102$.

(g) In 1966 at Berkeley, isotopes of element 102 from mass numbers 251–258 were reidentified accurately to lay to rest most of the discrepancies that had developed between the Dubna and Berkeley groups in energies, half-lives, and mass assignments.^{8,22} Some years later, in 1973, the longest-lived known nobelium isotope, 58-min

^{259}No , was discovered^{8,23} by R.J. Silva, P.F. Dittner, M.L. Mallory, and O.L. Keller, Jr., at the Oak Ridge National Laboratory, and Kari Eskola, Pirkko Eskola, Nurmia, and Ghiorso at the Lawrence Berkeley Laboratory.

(h) In 1967 at Berkeley, using the 3-min ^{255}No isotope that had been discovered at Dubna, the first chemical identification of nobelium was made when J. Maly *et al.*^{8,2} were able to perform aqueous solution chemistry to show that the 2+ state, *not the 3+ state*, of nobelium is the most stable in aqueous solution — a very important and rather unexpected finding. This result gave further proof that the original Stockholm experiments could not have been correct for chemical as well as for nuclear physics reasons. Nobelium (II) *could not have been eluted early* from a cation exchange resin column with ammonium α -hydroxisobutyrate, as Fields *et al.*^{8,1} had claimed.

There is a sequel to this work. During their experiments on the element 104 α -emitters, the Berkeley group discovered 1.8-s $^{250}\text{Fm}^m$. This low-lying isomer only manifested itself by transferring its 30-min $^{250}\text{Fm}^{gs}$ daughter when it decayed in vacuum. The question arose as to whether this mechanism could have played any role in the original Berkeley discovery experiments when ^{250}Fm was milked from ^{254}No after α -decay. After careful consideration, it was decided in the negative, for a number of reasons: (1) the observed milking half-life was 3 s, not 1.8 s; (2) the available recoil energy could only be eV rather than 100 keV; (3) the ^{250}Fm recoiling atom was probably not charged, since the element 102 milking experiment was performed at atmospheric pressure; and (4) Dubna, using a rare earth nuclide as a stand-in for their element 103 milking experiment, had shown that under similar conditions ec-daughters were transferred with an efficiency of less than 1%. For all of these reasons, Ghiorso *et al.* decided that their new, very unusual isomer could not have affected the conclusions of their milking experiment.

Though element 102 was originally named “nobelium” on the basis of the incorrect work at the Nobel Institute in Stockholm, it has been accepted by most of the scientific community and remains in

common use today. In 1967, after a hiatus of ten years, Ghiorso and Sikkeland,^{8,3} speaking for their collaborators at Berkeley, suggested that the name "nobelium" should be retained for element 102, along with its symbol, No. The name and symbol were reaffirmed by the IUPAC in its August 31, 1997, acceptance (Chapter 13, Table 13.5) of the previously approved names "mendelevium," "nobelium," and "lawrencium" for elements 101, 102, and 103.

8.3. Lawrencium (Element 103)

After the confusing discovery of element 102, which ended in disaster in 1959 with the destruction of the curium targets, the Berkeley group was extremely wary of repeating the same type of experiment very soon. In fact, the HILAC bombardment cave, which had been called by the ominous name HADES (Heavy Atom Detection Equipment Studio), was rebuilt to conform to much higher radiation safety standards, but was now labeled LIMBO (not an acronym!). The rationale here was that they were not confident yet that the new setup merited the name HEVN and so they compromised on LIMBO! (In Catholic theology Limbo is the way station between Heaven and Hell for those souls whose final destination has not been decided for reasons such as lack of baptism.)

Normally, to make element 103, the next step in their campaign, they would have used a large curium target again and bombarded it with ^{15}N ions. Ghiorso, together with Sikkeland, Robert M. Latimer, and Almon E. Larsh, decided, instead, to try their luck by bombarding small amounts of a mixed $^{249}\text{--}^{252}\text{Cf}$ target with ^{10}B and ^{11}B ions, for two reasons. First, the amount of radioactivity emitted by the target was more than an order of magnitude lower, and second, the cross section was predicted to be substantially larger than with curium as a target. In addition to that, the resulting nuclide that they would make would be heavier and, they assumed, longer-lived.

The 1961 Berkeley experiment was planned to be a very simple one. They would depend on nuclear physics to prove that they had found element 103, because there was as yet no adequate fast chemical

separation known that would do the job. This meant that they would use the well-established arguments of excitation functions, cross bombardments, and alpha systematics to identify anything that they observed. After they had found something they would reinforce it with some other technique.

To detect the α -particle radiations from element 103 they decided to take advantage of a very important new development that had just occurred at the Oak Ridge Laboratory (later to become the Oak Ridge National Laboratory). Cassimir J. Borkowski and J.L. Blankenship had recently demonstrated that silicon detectors could be used for measuring α -particle energies in unusual ways because of their small size and high resolution. Cass, one of the early members of the Ghiorso group at the Metallurgical Laboratory, had alerted Ghiorso to this new technique and suggested that he send his colleague Robert Latimer to Oak Ridge to learn how to make surface barrier silicon detectors.

In the meantime, at Berkeley, a suitable apparatus was designed and built to make use of this new detection capability. Recoils knocked out of the target were stopped in helium at atmospheric pressure and electrically directed to the surface of a thin copper tape. The tape was pulled periodically in front of a row of five of the new detectors to measure the α -spectra of the radiations emanating from the sequential spots. (See Fig. 8.4.) In this way energies and half-lives could be determined with fairly high efficiency and good resolution.

They were thwarted in their initial efforts, because tiny amounts of lead impurity in the target produced an overpowering background from α -emitters in the region just above lead on the nuclide chart. They had already encountered this problem in the work with the on-line grid chamber used in the second phase of the nobelium research, but this time it would plague them with a vengeance. This was because the weight of this californium target, the first ever to be bombarded, was a hundred times smaller than that of the lighter elements which they normally used, and this magnified accordingly the effect of the impurity for a given level of cross section. Harris and Latimer worked hard to reduce the lead level, but it seemed that no matter what they did, as fast as lead was removed from the target

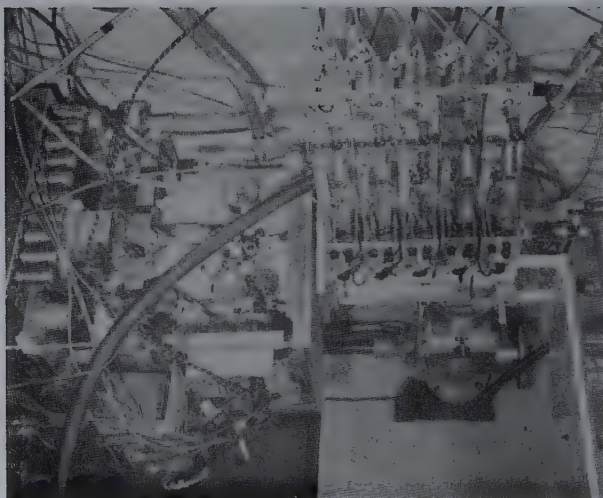
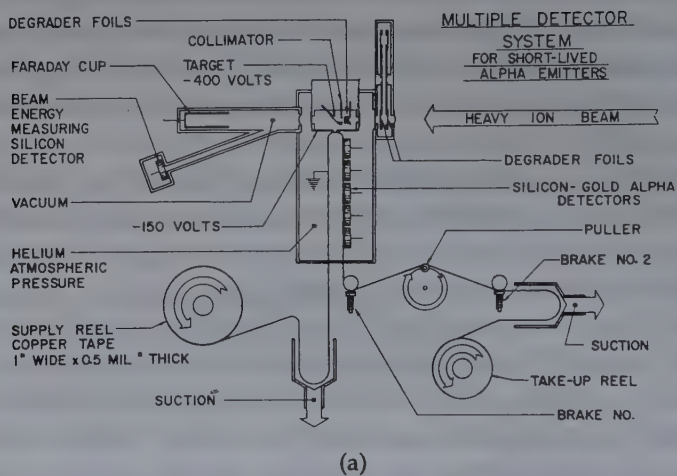


Fig. 8.4. Schematic (a) and photo (b) of the equipment used in the element 103 experiments.

somehow other sources of lead would find their way back in. They found that even the smog in the environment could contribute, so it too had to be excluded! They undertook a months-long effort to make the target free from lead using many different methods. They were handicapped by the fact that they had a limited amount of californium, otherwise they might have been able to vaporize a suitable target, perhaps separating the lead in the process.

Sikkeland and Ghiorso worked hard on the problem, too, but from a different perspective, and eventually they did find a better way. The idea was basically very simple. Why not make a final separation of lead *after* the target was made? The process worked on the principle that lead chloride was much more volatile than californium fluoride. All of the tiny amount of californium available was initially purified very carefully with ion exchange methods and then electroplated in the usual way as the hydroxide on the target substrate. This substrate was a 1-mg/cm² piece of nickel foil which had been first tested for lead by bombarding it in the HILAC beam. The electroplated californium hydroxide was converted to the fluoride in a wet HF environment and then subjected to HCl vapor to convert any residual lead to the chloride. The resultant mixture was carefully heated in vacuum to a temperature high enough to remove any traces of lead without vaporizing away the californium. This recipe was worked out very carefully and was found to accomplish the required purification, removal of the lead down to the level that was in the nickel itself, a very low level indeed. The final target of californium weighed only 3 µg, but was concentrated in a 0.051-cm² area. It had the following isotopic composition: 249, 3.3%; 250, 32.8%; 251, 12.3%; and 252, 50.8%.

This target was bombarded by ¹⁰B and ¹¹B ions from the HILAC, and the Berkeley experimenters were gratified to find a few counts at a new α-energy, 8.6 MeV, with a half-life of roughly 8 s. This was an activity that they had not seen before in bombardments of lead and they assumed that it was probably from element 103. To prove this assumption they decided that they would have to accumulate a large amount of data that would be unassailable and this proved to

be a very difficult and lengthy process. The bombardments were agonizingly slow in producing results, because the number of element 103 counts came in at a maximum rate measured in counts per hour. This made it exceedingly difficult to measure a good excitation function. The HILAC had just been upgraded to have a longer duty cycle which helped the experiment, but the 10-MeV/A boron beams still had to be degraded down to energies around 5 MeV/A. This process meant that a lot of power had to be dissipated in the aluminum foils used as degraders and it was often found that they had been partially melted by the concentrated beam. For some of the runs this meant that the energy at the target was very uncertain. In addition to this problem the final energy spread in the beam was made fairly wide by range straggling. This is the price that has to be paid when one is using a fixed energy accelerator. It would only be solved for the Berkeley experimenters when the variable energy SuperHILAC was built in 1971. The fact that there were four isotopes in the target also made interpretation of the excitation function exceedingly difficult. They trudged on, though, and finally convinced themselves that only an isotope of element 103 could explain their results.

As a part of the identification of the 8.6-MeV alphas, an extensive set of cross bombardments was made using ^{208}Pb , ^{209}Bi , ^{240}Pu , ^{241}Am , and ^{243}Am as targets and ^{12}C as well as ^{11}B and ^{10}B ions as projectiles. It was during some of these bombardments that they observed an 8.2-MeV activity with a half-life of around 15 s, which they decided was due to a new isotope of nobelium. This is discussed in the section on nobelium. By the conclusion of many runs they felt that they could assign the new 8.6-MeV α -activity to element 103 with complete confidence, but could not unequivocally assign its mass number. They felt that there was some evidence that it was 257, but that neither mass number 258 nor 259 could be excluded. Later work revealed that the α -energy was correct, a more accurate half-life was 4.3 s, and the mass number was 258. This first identification of an isotope of element 103 was published^{8,24} in 1961 by the discoverers, Ghiorso, Sikkeland, Larsh, and Latimer (Fig. 8.5). They suggested



Fig. 8.5. Berkeley discoverers of element 103.

that the element be named “lawrencium,” with the symbol Lw, in honor of Ernest O. Lawrence. The name was accepted by the IUPAC in 1971^{8,25} but the symbol was changed to Lr. The name and symbol were reaffirmed by the IUPAC in 1997.

A few years later, in 1965, the element was identified once again, this time genetically, by Donetz, Schegolev, and Ermakov^{8,26} at Dubna using the nuclear reaction $^{243}\text{Am}(^{18}\text{O}, 5n)$ to make a different isotope, ^{256}Lr . They used the same rotating wheel, double-recoil apparatus used in the experiments on ^{256}No to identify ^{252}Fm as its daughter by α -decay. In the case of ^{256}Lr , they chemically linked this same nuclide to ^{256}Lr using the genetic two-step (α -ec) decay sequence, $^{256}\text{Lr}(\alpha)^{252}\text{Md}(\text{ec})^{252}\text{Fm}$, the recoil milking indicating a half-life of ≈ 45 s for ^{256}Lr .

In spite of the short half-life of ^{256}Lr (now known to be 28 s), Silva, Sikkeland, Nurmia, and Ghiorso^{8,27} were able to establish in 1970 that in element 103, *in contrast to element 102*, the 3+ oxidation state is again the most stable in solution, as predicted by the actinide concept. These experiments required very difficult “atom-at-a-time”

fast chemistry, in which, on the average, only about one atom was detected in each of the several hundred manually conducted experiments.

Because both the Berkeley and Dubna groups contributed strongly to the correct identification of the isotopes of element 103 and their properties over the decade from 1961 to 1971, the TWG in 1992 (see Chapter 13, Table 13.1) recommended that credit for the discovery of lawrencium should be shared between them, and the name "lawrencium," symbol Lr, was reaffirmed.

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Chapter 9

Rutherfordium and Hahnium

9.1. Introduction

The history of the discoveries of elements 104 and 105, which here will be called "rutherfordium" (Rf) and "hahnium" (Ha), is one of decades of controversy and competing claims dating back to the period 1960–1977. Because of the difficulty in trying to assess the validity of the conflicting claims and suggestions for the names of these elements by researchers working at Berkeley in the USA and those at Dubna in the USSR, the International Union of Pure and Applied Chemistry (IUPAC) and its companion organization, the International Union of Pure and Applied Physics (IUPAP), agreed in 1974 to appoint an international *ad hoc* committee of neutral experts to include three each from the USA and the USSR and other countries (including the chairman), "to consider the claims of priority of discovery of elements 104 and 105 and to urge the laboratories at Berkeley (USA) and Dubna (USSR) to exchange representatives regarding these elements." The representatives from the USA were: Dr. Darleane C. Hoffman (then at the Los Alamos Scientific Laboratory), Dr. Earl K. Hyde (Lawrence Berkeley Laboratory), and Dr. O.L. Keller, Jr. (Oak Ridge National Laboratory). The representatives from the USSR were: Prof. V.I. Goldanskii (Institute of Chemical Physics, Moscow), Prof. S.P. Kapitza (Institute of Physical Problems, Moscow), and Acad. B.M. Kedrov (Institute of History of Science & Technology, Moscow). The representatives from neutral countries were: Chairman, Prof. J. Lewis (University Chemical Laboratory, Cambridge, UK), Prof. A. Baumgartner (Basel, Switzerland), and Prof. U. Stille (Braunschweig, Federal Republic of Germany).

Although the Committee never met, Earl Hyde visited the Chairman in Cambridge to discuss the matter and in 1975 the Chairman issued a request that the members of the Committee "prepare a draft of the history of the work and respective views in the naming of these elements by the Russian and American parties." Subsequent to this, the Committee carried out some informal activities to try to understand the problem and promote better communication and agreement between the Berkeley and Dubna groups, but it never finished its work nor prepared a report. However, the U.S. members of the Committee did decide to prepare a report to document the history of the discovery of elements 104 and 105, since they had already spent much effort to amass a comprehensive volume of literature on the subject including not only articles in refereed journals but reports of the various laboratories. This effort began in 1978–79 while Darleane Hoffman was on sabbatical leave at Berkeley, but was not completed until much later. It was first issued as a Lawrence Berkeley Laboratory (LBL) report, and then in 1987 it was published in *Radiochimica Acta*.^{9.1} The editors offered the Dubna researchers the opportunity to contribute a companion paper on the same subject, but they declined. For a detailed discussion of the claims to the discovery of elements 104 and 105 and a comprehensive bibliography, including reports of the various laboratories as well as all papers published in refereed journals, see Ref. 9.1.

9.1.1. *Rutherfordium (Element 104)*

The finding of the three US members of the 1974 *Ad Hoc* Committee, Hyde, Hoffman, and Keller,^{9.1} was, "We conclude that the information published in 1969 and 1970 by GHIORSO, NURMIA, HARRIS, ESKOLA, and ESKOLA^{9.2, 9.2a} was correct and that no other publications on the three isotopes of mass number 257, 259, and 261 preceded theirs. Their information fully meets the criteria for discovery of new elements stated by HARVEY *et al.*,^{9.3} and those stated by FLEROV and ZVARA.^{9.4} The earlier claims by the Dubna group for definitive identification of an isotope of mass number 260 based on

their results published in 1964 or in subsequent years up through 1969 cannot be accepted, nor do any of the results published later by this group reestablish the basis for their claim for priority of discovery. Therefore, we believe the Lawrence Berkeley Laboratory group should have the traditional right to suggest a name for the element. We suggest that their choice of the name rutherfordium, with symbol Rf, be adopted." (Note: The Dubna group had proposed the name "kurchatovium," with symbol Ku, for element 104.)

9.1.2. Hahnium (Element 105)

Concerning element 105, they^{9.1} said, "We conclude that the evidence presented by GHIORSO, NURMIA, ESKOLA, HARRIS, and ESKOLA^{9.5, 9.6} was definitive and convincing and that it fully meets the requirements of both statements^{9.3, 9.4} on *Criteria for Discovery of New Elements* which were cited in Ref. 9.1, Section VI. In particular, the proof of atomic number by genetic linkage to known isotopes of the daughter element 103 is very strong. Their work was performed and published at the same time as the Soviet work on spontaneous fission (SF) properties.^{9.7-9.11} We recommend that GHIORSO, NURMIA, ESKOLA, HARRIS, and ESKOLA be credited with the discovery of element 105 and be given the traditional right to name the element. We suggest that their choice of the name hahnium, with symbol Ha, be adopted." (Note: The Dubna group had proposed the name "nielsbohrium," with symbol Ns, for element 105.)

9.2. Review of 104 and 105 Discovery Claims

A brief description follows of some of the salient points concerning elements 104 and 105, some relevant background information on fission isomers, as well as some previously unpublished information about informal attempts by the 1974 *Ad Hoc* Committee to try to resolve the conflicting claims and achieve agreement on some of the major points of contention.

9.2.1. Fission Isomers

Several years before, when they first started using the apparatus described later, G.N. Flerov and his group had found a very prominent SF activity with a half-life of about 14 ms which they thought might be coming from an element 104 isotope. They soon found, however, that they could also make the same activity by bombarding the element 94 target with ions of lower Z than that of neon, whose Z is 10, so their usual rule, the Z of the target plus the Z of the projectile gives the Z of the final product, did not hold in this case. They had lowered the possible atomic number of the 14-ms activity down to element 97 or less when Ghiorso happened to visit Dubna in 1959. He, too, was immediately curious about its origin and he told Flerov that when he returned to Berkeley he also would do experiments to find out which element gave rise to this strange activity. Back at Berkeley he constructed a simple fission ion chamber with the target as one electrode to measure SF activity that decayed with a 14-ms half-life in the interval between the HILAC beam pulses. The target was also ^{242}Pu as in the Dubna experiments, and he immediately found that the 14-ms SF activity could be observed by bombarding the target with lighter ions than boron. By the time he had gotten down to using deuterium and other targets he had proved that it must be due to americium, element 95. Finally, the use of protons and an excitation curve proved that the 13.7-ms SF activity was in fact a new isomer of ^{242}Am and thus could be designated $^{242}\text{Am}^m$. This was a very unexpected discovery and Iz Perlman, who was very much interested in the matter, commented at the time that there must be something important that nature was trying to tell us — and he was absolutely correct! (Unfortunately, Ghiorso, preoccupied with the heavier elements, never took the time to publish his interesting discovery!) Earl Hyde happened to be making his own trip to Dubna at this juncture and Ghiorso, in a spirit of cooperation, told Earl to pass on this latest information to Flerov and his people. This came at the right time, for them because they had not yet determined the origin of the fissions themselves

and the information started Sergei Polikanov on the road to uncovering a whole new phenomenon, that of "shape isomerism."^{9.12} He and V.M. Strutinsky, who worked out the theory, would later receive the Bonner Prize of the American Physical Society for this seminal research. During the late 1960s and the early 1970s, a large number of additional fission isomers were found and characterized and the field was found to be quite extensive. Polikanov fell out of favor with Flerov later and it should be noted that Flerov is not a coauthor of the isomer paper, whose authors are S.M. Polikanov, V. Druin, V.A. Karnaukhov, V. Mikheev, A. Pleve, N. Skobelev, N. Subotin, G. Ter Akopian, and B. Fomichev!

Darleane Hoffman recalls that she was invited by Flerov to speak on their recent measurements at Los Alamos of neutron multiplicities of the heavy californium and fermium isotopes at the Conference on Interactions of Heavy Ions with Nuclei and Synthesis of New Elements (Fig. 9.1), held in Dubna in December 1977. The American



Fig. 9.1. Darleane Hoffman speaking at the Conference on Interaction of Heavy Ions with Nuclei and Synthesis of New Elements, Dubna, USSR, December 1977. Peter Möller is in the front row, center; Sergei Polikanov and Vilen Strutinsky are at the far right in the next row.

Physical Society then asked her to contact Polikanov and Strutinsky while she was in Dubna to notify them that they were to be corecipients of the Bonner Prize in nuclear physics, because letters and cablegrams to them had gone unanswered. This was at the height of the Cold War and she was to ask them if they could come to the US to receive the award, which included a cash prize, or if not, how they would like to receive their prizes. She had met Strutinsky many years earlier in 1966 at Lysekil, Sweden, where he spoke about his development of shell corrections to the liquid drop model which was instrumental in explaining the origin of fission isomers. She spoke there on attempts to produce elements beyond fermium in underground nuclear tests and had many discussions with Strutinsky on the implications of his calculations. Instead of the cash prize, he wanted one of the Texas Instruments hand-held calculators with the little magnetic strips on which programs could be stored. These little calculators were quite powerful for those times. In addition, he especially wanted to make sure he had enough of the paper tapes that went with it! So that was arranged through contacts in Copenhagen. Darleane was invited to the Polikanovs' apartment and enjoyed a delicious lunch with them. It became obvious that they were essentially under "house arrest" and only a short time later were finally able to leave the Soviet Union for CERN and later for GSI. It should be pointed out that the shape isomerism discovery was completely due to Flerov's attempt to make an isotope of element 104, so, although he never did find element 104 in this way he was indirectly responsible for a very important advance in nuclear physics.

9.2.2. Element 104

The original claim to the discovery of element 104 by G.N. Flerov's group at the Joint Institutes for Nuclear Research (JINR) at Dubna, USSR, was published in 1964.^{9,13} Utilizing the internal beam with a special apparatus (Fig. 9.2) placed inside the 300-cm heavy ion cyclotron at Dubna, they reported measuring an approximately

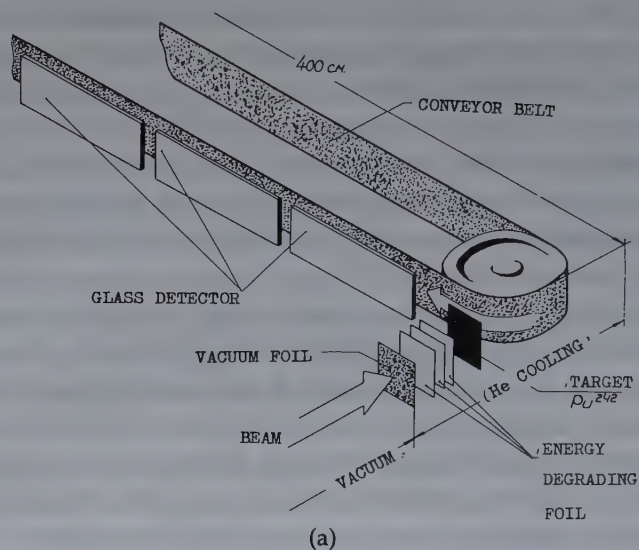


Fig. 9.2. (a) Schematic diagram of the apparatus used in Dubna to find the "0.3-s SF emitter" in the reaction of ^{242}Pu with ^{22}Ne projectiles. (b) Ivo Zvara, G.N. Flerov, and Al Ghiorso in a discussion in Prof. Flerov's office, Dubna, USSR, 1963.

0.3-s SF activity produced with a cross section of about 20 nb in bombardments of ^{242}Pu with ^{22}Ne . After performing a number of cross bombardments, they attributed it to $^{260}104$, assuming that it was produced via the $^{242}\text{Pu}(^{22}\text{Ne}, 4n)^{260}104$ reaction. Recoiling nuclei from the target were caught in a nickel conveyer belt which transported them past a series of phosphate glass fission detectors. The speed of the belt was varied over a wide range, depending on the half-lives to be expected, and the distributions of fission tracks in the detectors were analyzed to give the half-lives of the fissioning isotopes. They considered other assignments for this SF activity but, based on the dependence of its production cross section as a function of bombarding energy, they concluded that the $(^{22}\text{Ne}, 4n)$ reaction was most likely, that it was not consistent with a $(^{22}\text{Ne}, \alpha 4n)$ reaction, and thus could not be due to 3-s $^{256}102$, which was known to have a small SF branching ratio. A possible alternative assignment was to $^{259}104$, unknown at that time, which might be produced via the $(^{22}\text{Ne}, 5n)$ reaction.

In 1968, Donets and Shchegolev^{9,14} searched for an α -decay branch of the 0.3-s activity produced in the same reaction. They used an α -particle recoil transfer technique followed by a chemical separation to try to detect 22.7-h ^{252}Fm , the granddaughter of $^{260}104$. It was not detected and an upper limit of 20 nb was thereby set on the formation cross section for an α -emitting $^{260}104$. This would indicate that the α -decay branch for their hypothetical 0.3-s activity would be < 50%.

Although SF fragments can be readily and sensitively detected with fission track detectors, even when only a few events are recorded, only the half-life can be deduced; no information on the Z and the A of the fragments or the fissioning nucleus or information concerning mass-yield or kinetic energies of the fission fragments can be obtained. The fission process itself effectively destroys any direct information concerning the proton and mass numbers of the fissioning nucleus. In principle, it is possible to measure the proton number Z and mass number A of coincident primary fission fragments and in this way reconstruct the Z and A of the parent, but even today it has not been possible to perform the required

sophisticated measurements on the very few atoms of short-lived isotopes of new heavy elements which are produced. With presently available techniques a positive identification of the atomic number of a fissioning species is only possible by chemical separation or if the SF emitter is the daughter of a known α -emitter. Without that, identification is normally based on half-life systematics, excitation functions, cross bombardments, and other indirect determinations of atomic number. The systematics of SF half-lives known at the time of these discoveries^{9,15} were quite different from those based on our current^{9,16} experimental data (Fig. 9.3). It should be pointed out also that the recent identification (Fig. 9.4) of longer SF half-lives in the region of enhanced stability predicted by Sobiczewski *et al.*^{9,17} around the deformed shells at $Z=108$ and $N=162$ makes identification of SF nuclides based only on their half-lives still more difficult than was previously thought.

Differences in the measurement techniques employed by the Dubna and Berkeley groups should be pointed out. Rather than concentrating on SF detection, Ghiorso's group at LBL used various techniques based on the α -recoil phenomenon to separate and examine the subsequent decay of daughter products.^{9,2} The emphasis on SF measurements at Dubna in the early years to the near complete exclusion of α -particle measurements is perhaps understandable in view of the fact that Flerov and Petrzhak discovered the SF mode of decay (in ^{238}U) in 1940^{9,18} and it is an extremely sensitive method. In addition, solid state detectors and computer technology were generally more available in the USA than in the USSR.

Starting not long after the Dubna publication of their observation of an element 104 SF activity and not doubting the validity of their claim, the Berkeley team of Ghiorso, Matti Nurmi, James Harris, Kari Eskola, and Pirrko Eskola tried with great diligence to confirm the hypothesized 0.3-s SF emitter. The group fully expected that it would be able to do so and was appalled and frustrated when the initial experiments failed to find the activity. The question arose: Was the Dubna work in error or was the Berkeley work not sensitive enough to detect the new SF activity? After the Berkeley group had

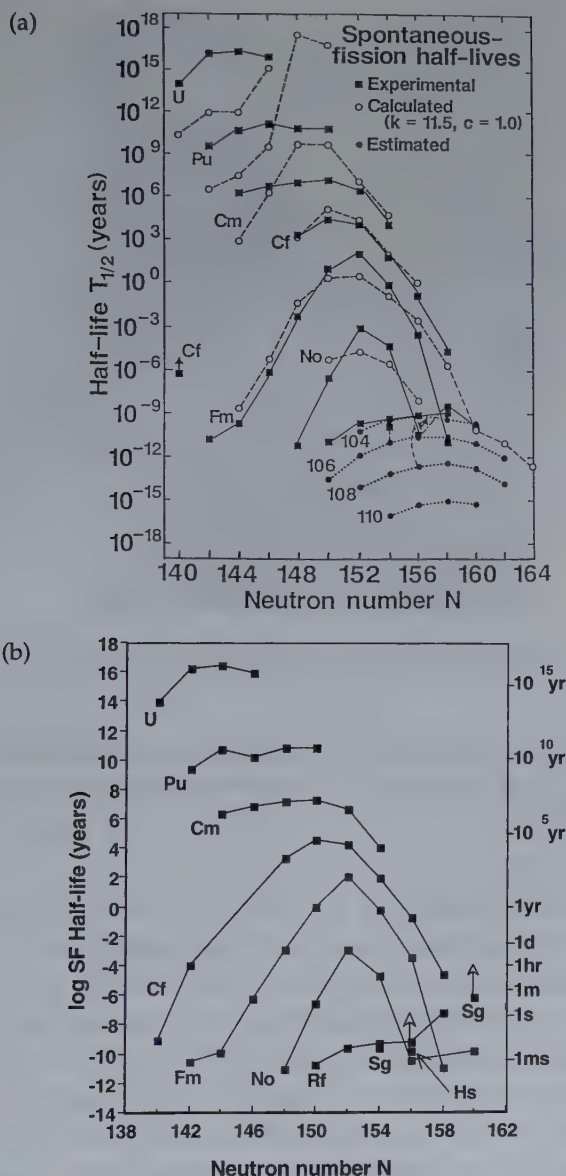


Fig. 9.3. (a) 1985 SF half-lives from L.P. Somerville *et al.*^{9.15} (b) Logarithms of known partial SF half-lives for even-even nuclei as of 1997. Arrows indicate lower limits. Adapted from Hoffman and Lane.^{9.16}

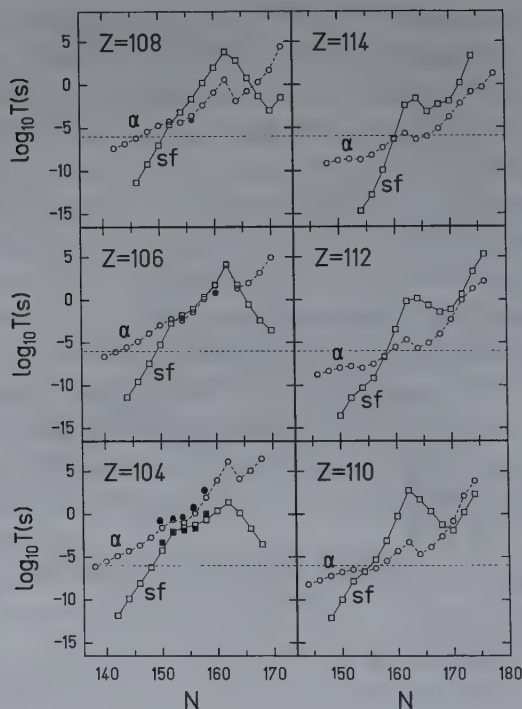


Fig. 9.4. Logarithms of calculated^{9,17} SF and α half-lives for even-even isotopes of elements 104–114. For comparison, experimental values are given by full points. Adapted from Hoffman and Lane.^{9,16}

demonstrated that there was no 0.3-s SF activity, Dubna discovered that the 0.3-s measurement was an experimental artifact due to fast neutrons inside the cyclotron. They now lowered the claimed half-life to 0.1 s, and this mandated that the Berkeley experiments be repeated on a faster time scale. Later yet, Dubna refined its experiment and, using an external beam, reported what seemed to be a precise 80-ms half-life. It was a very baffling experience for the Berkeley experimenters to find that their goal had become a moving target. Each time that new experiments at Berkeley showed that no such activity for element 104 existed, the Dubna team would counter with a new value or some new objection to the validity of the Berkeley



Fig. 9.5. Jim Harris with apparatus for electroplating targets, April 1969.

experiments. These will-o'-the-wisp chases covered a wide variety of target-projectile combinations^{9,2} at Berkeley. Einsteinium, californium, and curium targets (Fig. 9.5) were bombarded with various heavy ions from ^{11}B to ^{18}O that were calculated to give much higher yields and yet none of the SF half-lives, 0.3 s, 0.1 s, or 80 ms, were ever observed. In this process of searching, however, they did find a 20-ms SF emitter, which they deduced was most likely due to $^{260}104$.

These were strenuous experiments and went on sporadically for the next five years, always with the negative result that the claimed Dubna SF emitter could not be found even though the Berkeley group had more than adequate sensitivity. By 1967, the group decided that it would be more productive to concentrate its efforts on the new α -particle detection methods that they had developed during this time. They knew that the amount of information that would be obtained by finding an α -emitting isotope of element 104 would be

much more informative than an uncertain SF half-life. Ghiorso used to say that "one alpha particle was worth ten thousand spontaneous fissions." Of course, the exception to that rule occurs when the half-life of the SF emitter is long enough to allow chemistry to be used as an identifying tool as in the case of element 101 (see Chapter 7).

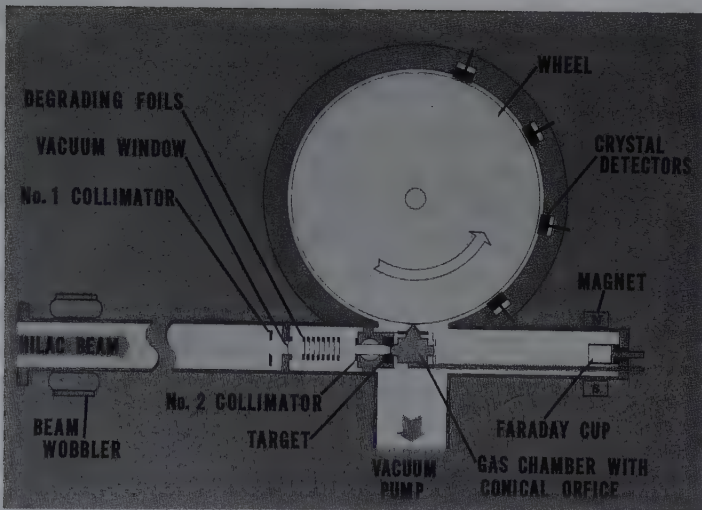
It took some time to develop the required technology to look for new and distinct α -particle emitters. The apparatus was based upon a vertically mounted wheel (Fig. 9.6), hence it became known as the VW, which acted as a stepped conveyor to carry recoil products sequentially to a series of high resolution gold surface-barrier silicon α -particle detectors for measurement of α - and SF-decay. The recoiling atoms were deposited by a helium gas jet from the target chamber onto the periphery of the wheel.

Early in 1968 enough ^{249}Cf became available to use in an attempt to produce element 104. The ^{249}Cf was obtained by β -decay from

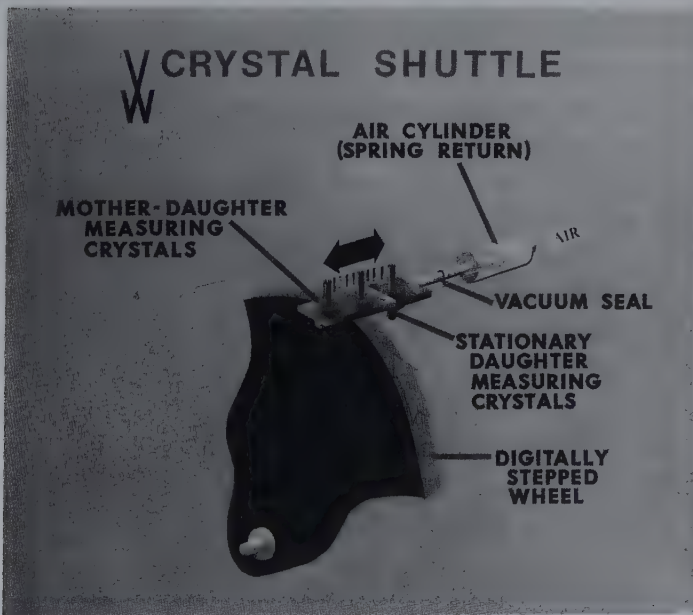


(a)

Fig. 9.6. (a) Al Ghiorso with original VW with only four detection stations, March 1964. (b) Schematic diagram of the first VW apparatus used for the discovery of element 104 α -emitters. (c) Detail of the VW crystal detector shuttle.



(b)



(c)

Fig. 9.6 (continued)

purified ^{249}Bk and a target with a total weight of 60 micrograms (about $200\text{ }\mu\text{g}/\text{cm}^2$) was prepared. At that time this was the entire world's supply of that pure isotope, the berkelium having been produced at the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory for the National Transplutonium Production Program under the direction of John Bigelow. John has played a vital role in the production of the actinide elements as targets for the transmutation of various transplutonium elements. The experiments at Berkeley, making use of the HILAC,^{9,2} were highly successful. Bombardments of the ^{249}Cf target with ^{12}C and ^{13}C produced the new α -emitters, $^{257}\text{104}$ ($T_{1/2} \approx 3.8\text{ s}$) with α -groups at 8.70, 8.78, 8.95, and 9.00 MeV and $^{259}\text{104}$ ($T_{1/2} \approx 3.4\text{ s}$) with α -groups at 8.77 and 8.86 MeV. The distinct alpha spectrum of each of the nuclides easily differentiated these new activities from minor lead-produced background effects. Although the circumstantial evidence was very strong that the new α -lines were emitted by isotopes of element 104, it was desirable that additional proof be obtained, and this was done in the following way.

To identify the mass and atomic numbers of these activities, the Berkeley group introduced an innovative method of α -particle recoil transfer to establish the genetic relationship between an unknown new isotope and its known daughter.^{9,2} At regular intervals, all of the detectors which viewed the wheel were shuttled off-line to positions opposite four similar detectors in order to measure the activity that had been transferred to the faces of the detectors by recoil due to those element 104 α -particles that had been emitted into the wheel. The energy and half-life of these α -recoil-transferred activities were measured in order to identify them as coming from known nobelium (102) daughter activities. Thus the link was made between the element 104 α -activities and their known daughter nuclides.

During the following year the isotope $^{261}\text{104}$ was discovered^{9,2a} by bombardment of ^{248}Cm by ^{18}O and found to have a half-life long enough for aqueous chemistry to be performed with it, as described in the next section.

9.2.2.1. Chemical Experiments

As mentioned before, Donets and Shchegolev^{9,14} of Dubna made early attempts to chemically measure and identify known daughter or granddaughter isotopes rather than the short-lived parent. However, the technique is not always applicable or as sensitive, is less direct, and depends on ensuring that the daughter species cannot be produced directly in the nuclear reaction used.

However, Flerov's group early realized that positive identification of a new element based only on its SF activity is very difficult and said in their original article^{9,13} that "...the authors believe it is quite desirable to conduct chemical experiments for additional identification as well as for the study of chemical properties of element 104." Ivo Zvara and his group at Dubna took on this formidable challenge of developing "one atom at a time" chemistry for a new element with unknown chemistry on the assumption that Seaborg's actinide hypothesis was correct and that the actinides ended with lawrencium (103) and, therefore, element 104 should be a heavy homolog of the group 4 elements zirconium and hafnium. They took on this task and developed gas-chromatographic separations for element 104 on the assumption that it was the first of the transactinide elements and should, therefore, exhibit properties similar to the group 4 elements zirconium and hafnium, and have volatile chlorides while the chlorides of the heavy actinides would have nonvolatile chlorides. Gas chromatography was selected because of its speed and good separation capability. Preliminary experiments conducted with the chlorides of the group 4 elements zirconium and hafnium, and some lanthanides gave promising results. Zvara's group pioneered gas phase studies of the transactinide elements and published detailed descriptions of their apparatus and experimental approach. In 1969 they reported^{9,19} that they had detected a total of 14 fission events in the hafnium fractions from a series of gas phase chromatographic experiments on element 104 produced via the $^{242}\text{Pu}(^{22}\text{Ne}, 4n)^{260}104$ reaction. They believed that the half-life could be determined if it had a value in the range of the transit time across the mica detectors

which was about 0.7 s, and they found that the distributions of fissions in the mica detectors was consistent with a half-life of 0.3 s. They also stated that the fission track distribution "shows that the effect was not caused to an appreciable extent by the decay of nuclides undergoing SF with half-lives of 0.014 s and 3.7 s." This, of course, ruled out contamination from 0.014 s $^{242}\text{Am}^{\text{m}}$ and ^{256}No . In a second set of similar chemical experiments, Zvara *et al.*^{9,20} stated that the distribution of fission tracks in the mica detectors "imply a half-life of somewhat less than one second," with a half-life of 0.5 s accepted as the correct value based on their complete set of data comprising some 67 events. Assuming a half-life of 0.5 s, they further calculated an adsorption enthalpy of 34 kcal/mol from their data, which correlated well with that for HfCl_4 . This was the only evidence given between 1966 and 1969 to verify that element 104 behaved as *eka*-hafnium in the chemical experiments. Later, in 1971, the Dubna group^{9,21} claimed to have achieved the chemical identification of element 104 using a new technique, gas thermochromatography; however, this work is subject to question, because the SF branching ratio for $^{259}\text{104}$, the isotope that they claimed to have observed, is too small for it to have been involved in their experiment. This was after the successful identification by the Berkeley group in early 1969 and 1970^{9,2} of 4.5-s $^{257}\text{104}$, 3-s $^{259}\text{104}$, and 65-s $^{261}\text{104}$.

In 1970, the first studies of the aqueous chemistry of element 104 were reported by Silva *et al.*^{9,22} at Berkeley using the known 65-s $^{261}\text{104}$ activity. These experiments showed that it was eluted from cation exchange resin together with tetravalent hafnium and zirconium tracers, well ahead of trivalent actinides including lawrencium (element 103). Figure 9.7 is a sketch of the chemical method used by Bob Silva, Jim Harris, and Matti Nurmi to perform this chemical separation. The element rutherfordium was positively identified by measuring the characteristic well-known α -decay of ^{261}Rf . These experiments established that it was a heavy member of group 4 in the periodic table and confirmed Seaborg's hypothesis that the actinide series did, indeed, end with element 103. Ron Loughheed reported at the 1975 Baden-Baden Transplutonium Element

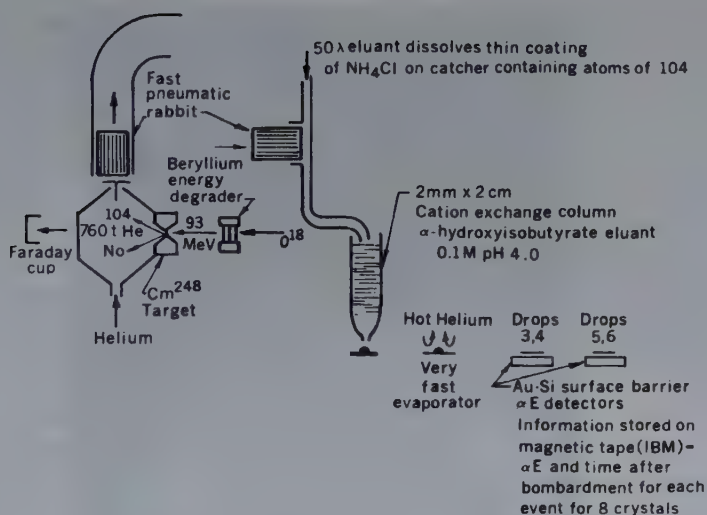


Fig. 9.7. Flow diagram for 104 chemistry experiments.

Symposium on studies of the chloride complexation of element 104 which he, Hulet, Nitschke, J.W. Wild, J.H. Landrum, and Ghiorso carried out^{9,23} using computer automation to perform rapidly all the chemical manipulations, to prepare the α -sources and to perform α -spectroscopy. A photo of this automated system, called the Fast Automatic Kjemistry Experiment, or FAKE, is shown in Fig. 9.8. More than 100 experiments were conducted with reversed-phase chromatography columns consisting of trioctylmethyl ammonium chloride sorbed on an inert support. The results showed that in concentrated HCl Rf behaves like the group 4 elements and forms strong anionic chloride complexes which extract onto the columns while the trivalent actinides form weaker complexes which do not extract. The Rf and group 4 elements Zr and Hf were then eluted together with 6 M HCl, indicating the conversion to neutral or cationic species. No further detailed studies comparing the behavior of rutherfordium with its lighter homologs were performed until the late 1980s.



Fig. 9.8. Mike Nitschke with his invention, the Fast Automatic Kjemistry Experiment (FAKE), for performing chemistry on element 104.

9.2.2.2. *Confirmation of the Berkeley Discovery*

A decisive confirmation of the original 1969 claim by the Berkeley group to the discovery of 4.5-s $^{257}_{104}$ produced in the $^{249}\text{Cf}(^{12}\text{C}, 4n)$ reaction was made at the Oak Ridge National Laboratory in 1973 by Bemis *et al.*^{9.24} They measured the X-ray spectrum in coincidence with α -particles attributed to $^{257}_{104}$ and showed that the X-rays were characteristic of the energies and relative intensities of the K X-rays of element 102, the α -decay daughter of element 104. Much later, in 1985, Hessberger *et al.*^{9.25} produced $^{257}_{104}$ via the $^{208}\text{Pb}(^{50}\text{Ti}, 1n)$ reaction at the UNILAC in Darmstadt, and measured a half-life and α -particle spectrum similar to those originally reported by Ghiorso *et al.*^{9.2}

9.2.2.3. Attempts to Reconcile Measurements of the SF Half-Life of $^{260}104$

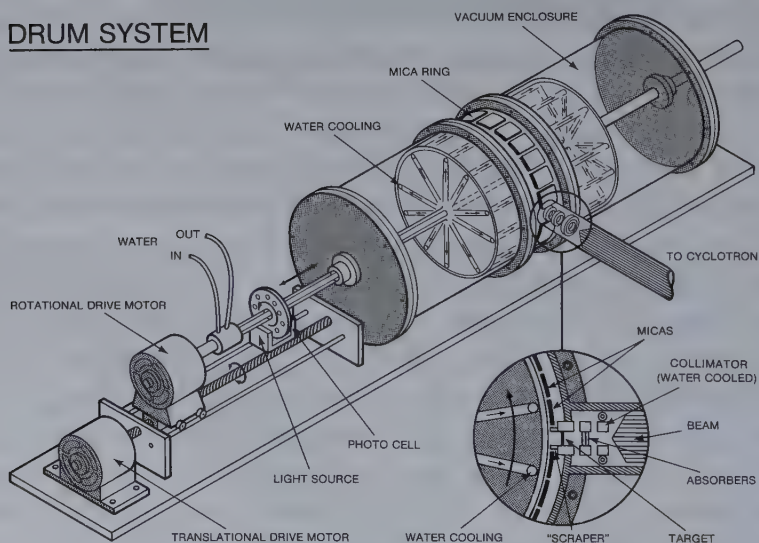
In 1975, Druin *et al.*^{9,26} studied the reaction of a ^{246}Cm target with ^{18}O projectiles and found an SF activity with a half-life of 80 ± 20 ms which was produced with a 1.5-nb cross section. Based on the excitation function and angular distribution, they assigned it to $^{260}104$ produced by the (^{18}O , 4n) reaction. These results contradicted the negative results reported in 1969 by Ghiorso *et al.*^{9,2} for this reaction, and stimulated (Fig. 9.9) the Berkeley group in April 1976 to restudy the ^{249}Bk (^{15}N , 4n) $^{260}104$ reaction,^{9,27, 9,28} which was expected to have the best cross section, of the order of 25 nb. Scientists from four American laboratories (LASL, LLNL, ORNL, LBL) and Ivo Zvara, an observer from Dubna, were present for these experiments. A large metallic drum rotating at high speed which could also be moved laterally was used (Fig. 9.6) to collect the recoiling products, and a series of mica sheets served as fission fragment detectors. Several bombarding energies were tried — but no trace of an 80-ms activity was seen although SF activity with a half-life of 20 ms and a peak cross section of about 19 nb was found! Ghiorso presented a preliminary report on these results^{9,28} at the 3rd International Conference on Nuclei Far from Stability, held at Cargèse, Corsica, France, in May 1976. They were sharply criticized by the Russians.

As one of the informal activities of the IUPAC Committee, a meeting of some of the principals was arranged during this international conference. It was organized and chaired by Darleane Hoffman and Günter Herrmann (University of Mainz and GSI, Germany). Among those present were: Albert Ghiorso (USA), G.N. Flerov (USSR), Mike Nitschke (USA). It was an extremely heated meeting. A. Zylic (Poland) served as interpreter for the meeting. Ghiorso reiterated his 1969 claim^{9,2} that his group was unable to confirm the existence of a 0.3-s isotope of $^{260}104$ or $^{259}104$. No evidence for this activity could be found in the reaction $^{246}\text{Cm}(^{18}\text{O}, 4\text{n})$ or $^{248}\text{Cm}(^{16}\text{O}, 4\text{n})$. It was then agreed that the USA and USSR would exchange scientists and attempt to reproduce the reactions and claims of each group. As a



Fig. 9.9. Photo taken on September 23, 1975, during the visit of Ghiorso and Seaborg to the Laboratory of Nuclear Reactions, Dubna, USSR, to discuss experiments to try to reconcile conflicting measurements of the SF half-life of $^{260}_{104}$. Flerov also made the surprise announcement of the discovery of element 106 at this meeting, later proven to be erroneous (see Chapter 10). Left to right: V.A. Druin, G.T. Seaborg, A. Ghiorso, G.N. Flerov, Yu. Oganessian, I. Zvara.

DRUM SYSTEM



XBL 7912-13728

Fig. 9.10. Drum detection system.

beginning, Flerov then invited Berkeley to send a representative to Dubna to participate in experiments to measure the SF activities produced in the reaction $^{242}\text{Pu}(^{22}\text{Ne}, 4n)^{260}104$. This was the original reaction in which Flerov's group had claimed discovery of element 104 in 1964^{9,13} based on the detection of a 0.3-SF activity produced with a cross section of about 20 nb.

A few months later, Mike Nitschke (LBL) did go to Dubna to participate in experiments there between August 30 and September 20, 1976. According to Mike's trip report,^{9,29} the Dubna group agreed, much to his surprise, that the $^{249}\text{Bk}+^{15}\text{N}$ reaction would be investigated first in order to compare directly with the earlier experiments conducted at Berkeley in which a 20-ms, but no 80-ms activity, was found. But, unfortunately, due to an earlier accident, a hole had been burned in the ^{249}Bk target and 2/3 of it was lost, so the first reaction to be studied was $^{246}\text{Cm}(^{18}\text{O}, 4n)$ to try to make the SF activity variously reported by Dubna as 300, 100, and finally 80 ms. And, indeed, according to Mike's report, a 19 ± 2 ms activity was found, in complete agreement with the Berkeley results from April 1976. Closer analysis showed that it could be decomposed into a short-lived activity (14 ms) and a longer-lived one. Determination of the cross section was difficult, because the beam intensity readings were not reliable, but the best estimate was $1.5 \text{ nb} \pm 50\%$. The second set of experiments on this reaction was similar, but instead of 8 m, the total tape length was 45 m to spread out the long-lived background activity so it did not interfere with the determination of the shorter half-lives. And now, the speed was set at only 70 cm/s instead of 270 cm/s, but because of the low speed and a detector-free zone between the point where the recoils were deposited on the tape and the first glass fission detector, it was impossible to properly account for the influence of short-lived activities. This "dead-time" of about 50 ms corresponded to more than three half-lives for the $^{242}\text{Am}^m$. After two weeks of these experiments, a target of ^{249}Bk was finally available. It was 0.6 mg/cm^2 thick, but contained only 0.18 mg/cm^2 of ^{249}Bk , the rest being impurities! The chemists had experienced great difficulty in purifying the ^{249}Bk even though

A.A. Kosyakov had come from Moscow to help and Yu. S. Korotkin had been ordered back from vacation to electroplate the target. The first berkelium experiment with the fast 8-m system showed a short-lived SF activity in the range of 20 ms, and a large background from 2.6-h ^{256}Fm . This prevented any possible identification of an ~100-ms component, and the experiment was then repeated with the 345-m tape system. In two such runs, an activity of about 100 ms with a 20–30 ms uncertainty was found — the cross section for ^{256}Fm was higher than in Berkeley. It was estimated that the cross section for the 100-ms activity was as low as 2 nb. An additional experiment with the berkelium target was conducted at a higher beam energy where the (^{18}O , 4n) reaction should be reduced and a shorter half-life of about 45 ms was observed. The shorter half-life was qualitatively explained by the larger influence of the short-lived component, either 14 or 20 ms. Mike says, "During my trip to Dubna I was not able to come to a final resolution of the 104 question." He concluded that an experiment must be performed which was designed to measure short-lived activities (~20 ms) and simultaneously determine the amount of long-lived activities (~100 ms) under low background conditions using a target of high purity. At the conclusion of his stay he met with Yu. Oganessian and V. Druin to discuss future experiments in both laboratories. He agreed to try to do a low background experiment in Berkeley. Druin and coworkers would continue their experiments with curium and berkelium targets with the recommended improvements for at least three months at the U-200 cyclotron. They were to report their results, together with results from previous experiments, at the December 1976 heavy element meeting in Dubna. Near the end of Nitschke's stay at Dubna, G.N. Flerov convened a meeting with about 25 scientists for a discussion of the results obtained. Flerov and Zvara criticized the 1968–69 Berkeley experiments, but agreed that the 1976 data were of "good quality." Druin presented an objective comparison between the Berkeley and Dubna equipment. After this Mike had about an hour to present the Berkeley results. The final discussion centered mainly on questions of background

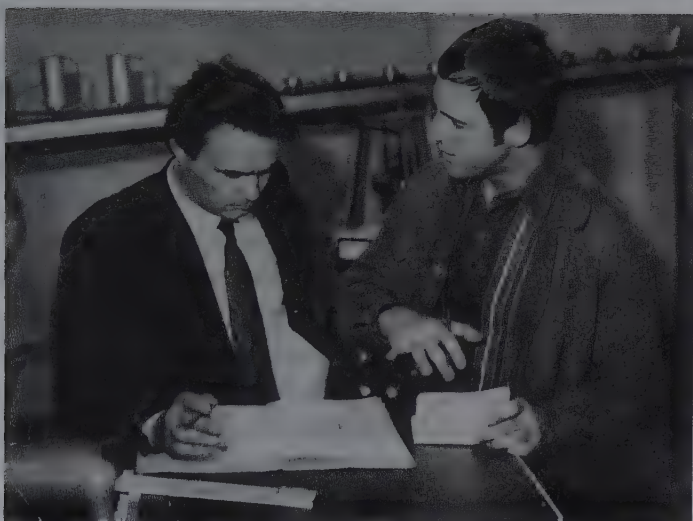


Fig. 9.11. E.D. Donets and Mike Nitschke in later discussions at Dubna, 1990.

and the upper limit set for the production of $^{260}\text{104}$ in the berkelium experiments.

Analyses of the data from the last two berkelium runs at Dubna were made later by groups at both LBL and Los Alamos. Neither group was able to identify an 80-ms activity in their least-squares analyses of the data, but an approximately 20-ms activity was found if proper consideration of a long-lived fission component (presumably ^{256}Fm) was included in the least-squares analysis.

In 1981 Nitschke *et al.*^{9.27} published a more complete review of the Berkeley search for the elusive 80-ms SF activity in bombardments of ^{249}Bk with ^{15}N . A cross section limit of 0.5 nb was set for the 80-ms activity, but a 23 ± 2 -ms activity with a maximum cross section of 19 ± 4 nb at 82 MeV was found. This SF activity was distinct from the 14-ms $^{242}\text{Am}^{\text{m}}$ and was consistent with assignment to $^{260}\text{104}$.

In later experiments, Somerville *et al.*^{9.15} used a 1000-m-long metal tape system with mica SF detectors to study half-lives ranging from milliseconds to hours (Fig. 9.7). Their studies of the $^{249}\text{Bk} + ^{15}\text{N}$ and $^{248}\text{Cm} + ^{16}\text{O}$ reactions showed no evidence for the 0.3-s, 0.1-s, or

80-ms activities reported by the Dubna group in these reactions. However, they did find clear evidence for an approximately 20-ms activity in these reactions and other reactions and concluded that 20 ± 1 ms was the best half-life for $^{260}_{104}$. Dubna physicists in a 1985 publication^{9,30} reported a value of 28 ± 6 ms, closer to the LBL value than the previous 80-ms Dubna value.

In the early Dubna chemical experiments, a half-life as short as 20 to 30 ms could not have been detected. Furthermore, since calculation of the heat of adsorption depended on the use of a 0.5-s half-life, which was wrong, the calculation that the heat of adsorption was similar to that of HfCl_4 was also wrong. Therefore, the chemical experiments of 1966–69 could not be considered as confirming the assignment of the initially observed 300-ms or later 80-ms or 20-ms SF activity to element 104 based on the similarity of its chemical properties to hafnium.

9.2.3. *Element 105*

Results of a preliminary search for α -emitting isotopes of element 105 from the $^{243}\text{Am}(^{22}\text{Ne}, 4 \text{ or } 5n)$ reaction were described in an unpublished 1968 Dubna report.^{9,31} A new system was used in which recoiling reaction products were transported in a N_2/He gas mixture via a capillary through the central hole of an annular silicon semiconductor detector. They were collected on the surface of a metal wheel and the α -particles were detected with the silicon detector. However, minute amounts of lead impurity in the target tended to mask the energy region of interest for $^{260,261}_{105}$. Attempts were made to perform time correlation measurements with their $^{256,257}\text{Lr}$ daughters, both believed at that time to have 35-s half-lives. (The half-life of the mass number 257 isotope was later measured to be 0.7 s, which changed the conclusions of the study.) When a start pulse in the region of 8.8–10.3 MeV was detected, the beam was interrupted for 160 s to wait for registration of α -particles from the daughters. After analysis of the data, the results were taken as tentative evidence that α -particles with energies from 9.1 to 9.8 MeV

(weak peaks at 9.15, 9.4, and 9.7 MeV) were emitted by isotopes of element 105 with mass numbers 260 and 261. Additionally, they concluded that the half-life for the 9.4-MeV α -emitter tentatively assigned to $^{261}\text{105}$ was in the range of 0.1–0.3 s; the 9.7-MeV α -emitter was tentatively assigned to $^{260}\text{105}$, which had a formation cross section of 0.1 nb and a half-life > 0.01 s. Because of the smaller-than-expected cross section, subsequent experiments^{9,32} were undertaken in 1970 to look for SF activities using the same apparatus as for the element 104 experiments. They found an SF activity with a half-life of 1.8 ± 0.6 s, and based on angular distribution and excitation function measurements, attributed the activity to either mass 260 or 261, but with a preference for mass number 261 of element 105. The authors stated that additional and unambiguous identification that an isotope of 105 had been produced would be made later by high speed radiochemical studies. Druin *et al.*^{9,10} again studied the α -decay and concluded that they had seen groups at 9.1 and 8.9 MeV which decayed with a 1.4-s half-life, but their energy resolution was poor and their spectrum in the 8.8–9.2-MeV range shows little resemblance to the much better data published at the same time or earlier in 1970 by Ghiorso *et al.*^{9,5} and confirmed in 1977 by Bemis *et al.*^{9,33} Results were consistent with the earlier study and the half-life of about 1.5 s was consistent with the SF measurements and appears to belong to the same isotope of element 105. However, Druin *et al.*^{9,10} observed a 35-s half-life, which they attributed to the $^{256,7}\text{Lr}$ daughters. Since it was later shown that the half-life of ^{257}Lr is 0.7 s, it would indicate that they had produced the isotope of element 105 with mass number 260 rather than 261. Based on current knowledge of SF half-life systematics and hindrance factors for odd–odd nuclei, this isotope is even less likely than mass number 261 to have a significant SF branch.

In early 1970, Ghiorso *et al.*^{9,5} reported the production of $^{260}\text{105}$ via the $^{249}\text{Cf}(^{15}\text{N}, 4n)$ reaction at the heavy ion linear accelerator (HILAC) at LBL. It α -decayed with a half-life of 1.6 ± 0.3 s. They measured α -groups of 9.06 (55%), 9.10 (25%), and 9.14 MeV (14%) and set a limit of 20% on its decay by SF. Its A and Z assignment were unequivocally

assigned based on the measurement of its genetic relationship to its known 30-s ^{256}Lr daughter. Reaction products recoiled from the thin target into helium gas which was pumped through a small orifice into a vacuum chamber, where the reaction products were deposited on the periphery of the VW, which was periodically rotated into positions next to a series of silicon solid-state detectors to measure the α -particle energies. Approximately half of the 105 daughter activities recoiled into silicon detectors, and each of these primary detectors was periodically shuttled into position opposite similar detectors for analysis of the daughter α -activities. This complex system involved 20 detectors, and a computer was used to tag and record the spectra from each detector. The daughter spectrum and half-life were consistent with the known spectrum and half-life of ^{256}Lr . The excitation function also agreed with the yield curve calculated for a (^{15}N , 4n) reaction.

A year later, in 1971, Ghiorso *et al.*^{9,34} used similar techniques to identify two new isotopes of element 105, 1.8-s $^{261}105$ (8.93-MeV α -group) and 40-s $^{262}105$ (8.45-MeV α -group) in bombardments of



Fig. 9.12. Discoverers of elements 104 and 105, March 1969. *Left to right:* Matti Nurmia, Jim Harris, Kari Eskola, Pirkko Eskola, Al Ghiorso.

^{250}Cf with ^{15}N ions and ^{249}Bk with both ^{16}O and ^{18}O ions. Genetic linkages and α -recoil correlations with the known lawrencium daughter activities were used to confirm the assignments. A photo of the discoverers of elements 104 and 105 is shown in Fig. 9.12. In 1977, Bemis *et al.*^{9,33} completely confirmed the LBL results for $^{260}\text{105}$ and provided additional proof of the assignment to element 105 by the measurement of the L-X-rays from the daughter Lr (103) in coincidence with the 1.5-s $^{260}\text{105}$ α -activity, and by establishing the time correlation between the 1.5-s $^{260}\text{105}$ α -activity and its 26-s ^{256}Lr daughter α -activity Bemis and coworkers further stated that they saw no evidence of the 8.9-, 9.4-, or 9.7-MeV α -groups reported by the Dubna groups^{9,10, 9,31} and that "The unique identification provided for element 105 in our present experiments unequivocally supports the discovery claims for element 105 proffered by Ghiorso *et al.*"

9.2.3.1. Chemical Experiments

In JINR internal reports, Zvara *et al.*^{9,35, 9,36} described their experiments to investigate the chemical properties of the 2-s SF activity reported by Flerov *et al.*^{9,31} They proposed that the chemical properties of element 105 must be analogous to tantalum and that its higher valent chloride (and possibly its oxychloride) must be rather volatile. Because of the short half-life, the recoiling atoms were slowed down in a stream of hot (300°C) nitrogen and transported away from the reaction chamber and mixed with chlorinating agents such as thionyl chloride or thionyl chloride and TiCl_4 . This gas stream then moved through a 170-cm-long glass tube, with the first 30 cm held at 300°C to remove nonvolatile chlorides. In the second part, the temperature gradient dropped to 50°C in order to deposit chlorides of different volatilities at different positions along the tube. Mica plates for SF detection were placed inside this part of the tube. After calibration with a number of fission product and heavy element chlorides, a series of experiments with the reaction of ^{243}Am with ^{22}Ne ions was performed. (^{170}Hf was produced from a thin coating of samarium on the target to monitor and calibrate the operation of the system.) The

18 SFs which were detected were located near where the ^{170}Hf was deposited, at a higher temperature than where ^{90}Nb was deposited. Although the authors concluded that the SF activity came from an element forming a comparatively volatile chloride as might be expected for eka-tantalum (element 105), and that the observed pattern of the fission tracks might have been distorted by decay of the short-lived species during transport through the tube and thus indicate a volatility lower than the correct one, the results are somewhat less than convincing. Certainly, they do not constitute proof that the 2-s SF activity can be attributed to element 105.

Later experiments with bromides and a nickel tube, reported in 1976,^{9,37} again show that the SF activity, although more volatile than the actinide bromides, seems to deposit over the same region as hafnium, but not in the same region as niobium. If the tracks actually were from element 105, then the technique does not have high enough resolution to distinguish between elements 104 and 105 and cannot constitute proof that element 105 was observed. In any case, these experiments were much too late to help substantiate the claims to the discovery of element 105.

The long debate over the priority of the discovery of elements 104 and 105, the ensuing naming controversies, and the role played by the Transfermium Working Group (TWG) appointed in 1987 by the IUPAP and the IUPAC to try to resolve these issues are discussed in detail in Chapter 13.

9.3. Recent Chemical Studies of Rutherfordium and Hahnium

The gas phase studies of the halides of elements 104 and 105 in the 1960s described earlier indicated increased volatilities compared to the actinides and suggested that they were probably members of groups 4 and 5, respectively, of the periodic table. However, these studies depended on interpretations and models based on the half-lives of the observed SF activities. Positive identification of the 104 and 105 nuclides under investigation was not made, and detailed

studies of the differences in their behavior relative to their lighter homologs in groups 4 and 5 were not possible.

No investigations of the solution chemistry of hahnium, nor additional detailed comparisons of the behavior of rutherfordium relative to zirconium and hafnium, were performed prior to the late 1980s. About that time, renewed interest began in performing more detailed experimental studies of the chemical properties of the heaviest elements in order to compare them with those of their lighter homologs in the same group. The major impetus for this was the prediction by Keller^{9,38} that because of relativistic effects rutherfordium might have the valence electron configuration $7s^27p^2$ rather than that of $6d^27s^2$, which would be predicted by analogy to the $5d^26s^2$ configuration of its lighter homolog, hafnium. Similarly, hahnium might have a $7s^26d7p^2$ configuration rather than the $6d^37s^2$ configuration analogous to that of $5d^37s^2$ of its lighter homolog, tantalum. In fact, if the $7s^2$ electrons were sufficiently stabilized due to relativistic effects, it was even conceivable that hahnium, unlike its lighter group 5 homologs, might exhibit a stable 3+ rather than a 5+ oxidation state in aqueous solutions.

The reactions now typically used to produce the longest-lived isotopes of the transactinide elements 104 and 105 for chemical studies are $^{248}\text{Cm}(^{18}\text{O}, 5n)^{261}\text{Rf}$ (78 s) and $^{249}\text{Bk}(^{18}\text{O}, 5n)^{262}\text{Ha}$ (34 s). The reaction products recoiling from relatively thin ($< 1 \text{ mg/cm}^2$) targets are transported from the reaction chamber to a suitable collection station or instrument outside the accelerator. The reactions to produce ^{261}Rf and ^{262}Ha have cross sections of the order of 5 nb, which results in the production of about two atoms per minute, but after the transport efficiency, chemical yield, detection efficiencies, and 50% decay are folded in, only about 0.1 α -decay per minute is actually detected.

The first investigations of the aqueous chemistry of element 105 to determine if it, indeed, behaved as the heaviest member of group 5 of the periodic table (Fig. 1.6) were not conducted until 1987. By that time, the nuclear properties of 34-s ^{262}Ha and its α -decay daughter,

4.3-s ^{258}Lr , were well known. This meant that ^{262}Ha could be used in chemical experiments and positively identified as element 105 via the established method of α - α correlations. Hoffman recalls that they had sufficient quantities of the required 320-d ^{249}Bk (received from the US-DOE-funded transplutonium element production program at the Oak Ridge National Laboratory, a program started by Seaborg and Ghiorso in the 60s!) to make the target. So the question was: What chemistry could be manually carried out quickly enough to separate element 105 that would also indicate whether it behaved as a heavy homolog of the group 5 elements niobium and tantalum? They both exhibit the (V) oxidation state, and probably exist in aqueous solution as hydrolyzed or complexed species. About this time, calculations of relativistic effects suggested that stabilization of low spin electronic orbitals might even be strong enough to allow the existence of Ha^{3+} in solution, so first experiments were aimed at simply finding out whether the (III) or (V) oxidation state was most stable in aqueous solution.

In the Spring of 1987, Günter Herrmann, of the University of Mainz, Germany, was in Berkeley as a Visiting Miller Research Professor; Darleane Hoffman was his Faculty Host, so he was closely associated with her group's research. She remembers: "He suggested that we try to adapt some of the fast chemistry that had been used in fission product separations for niobium at Mainz. Both niobium and Pa (V) were known to sorb on glass from strong nitric acid and it was presumed that tantalum and maybe element 105 would also. Ken Gregorich, then a postdoc in my group, worked out what we called 'the glass chemistry,' which was performed on ^{262}Ha (Fig. 9.13) produced at the 88-Inch Cyclotron at the Lawrence Berkeley Laboratory (LBL). Our group (mostly graduate students) performed some 801 manual separations on element 105 taking 50 s each, which showed^{9,39} that hahnium sorbed on glass from strong nitric acid like niobium, tantalum, and protactinium, but unlike the group 4 elements zirconium and hafnium, or heavy trivalent actinides, which do not sorb. The energy and time distribution of the α -decays of ^{262}Ha and



Fig. 9.13. Darleane Hoffman and Ken Gregorich with a simple setup for the first aqueous chemistry experiments on element 105.

its daughter, ^{258}Lr , were recorded. The α - α correlations between them provided the proof that we were measuring hahnium, so we were *not using an unidentified element to try to determine unknown chemical properties!* We were exceedingly excited to discover when we analyzed our data that the simple 'glass chemistry' sorbed hahnium extremely well. Then, at the end of the hahnium experiments, we performed exactly the same chemistry on isotopes of niobium, tantalum, and the group 4 elements zirconium and hafnium, also produced on-line at the Cyclotron, and confirmed that these lighter group 5 homologs sorbed while the lighter group 4 elements did not. Furthermore, analysis of our hahnium samples also showed that fermium and other trivalent actinides produced in the reaction did not sorb. So we were now quite sure that hahnium was behaving like a group 5 element in the (V) oxidation state.

“Encouraged by this success, we decided to try extractions of the group 5 elements into methylisobutyl ketone (MIBK) from mixed nitric/hydrofluoric acid solution. Under these conditions, tantalum is known to extract while niobium remains in the aqueous phase. Much to our astonishment, hahnium *did not extract into the MIBK with tantalum* as we expected, but remained in the aqueous phase with niobium! This unexpected difference in extraction behavior between hahnium and tantalum launched us on what was to be a very long and fruitful program to study the chemical properties of the transactinide elements in more detail to try to assess the possible influence of relativistic effects on their chemistry. Our colleagues from GSI, Darmstadt, and the University of Mainz, Germany, and from the Paul Scherrer Institute and the University of Bern, Switzerland, expressed interest in coming to Berkeley to collaborate with our group in starting these detailed studies of the chemistry of hahnium. (These groups had first brought automated systems to LBL in 1986 to use in collaborative studies of both the aqueous and gas phase chemistry of lawrencium at the 88-Inch Cyclotron.)”

The first element 105 collaboration began at LBL in 1988 when the GSI–Mainz groups under the leadership of Jens Kratz and Matthias Schädel brought the Automated Rapid Chemistry Apparatus (ARCA II) (Fig. 9.14)^{9,40} to conduct a series of chromatographic column separations on element 105 and its homologs. The PSI–Bern groups led by Heinz Gägeler brought the On-Line Gas Chemistry Apparatus (OLGA) (Fig. 9.15), a continuous on-line isothermal gas chromatography system developed^{9,41} at the Paul Scherrer Institute in Switzerland for studying the volatility of the halides of hahnium and its homologs. An instrument (Fig. 9.16) similar to OLGA, the Heavy Element Volatility Instrument (HEVI), developed later by Kadkhodayan *et al.*^{9,42} at LBL, was also used to perform isothermal chromatographic gas phase studies of the volatilities of the halides of rutherfordium and hahnium. Subsequent collaborative investigations to study hahnium chemistry were hosted at Berkeley in 1990, 1993, and 1997. A photo of the 1990 collaboration which also includes Ivo Zvara from Dubna is shown in Fig. 9.17.



Fig. 9.14. Jens Kratz and Matthias Schädel with ARCA, 1988.

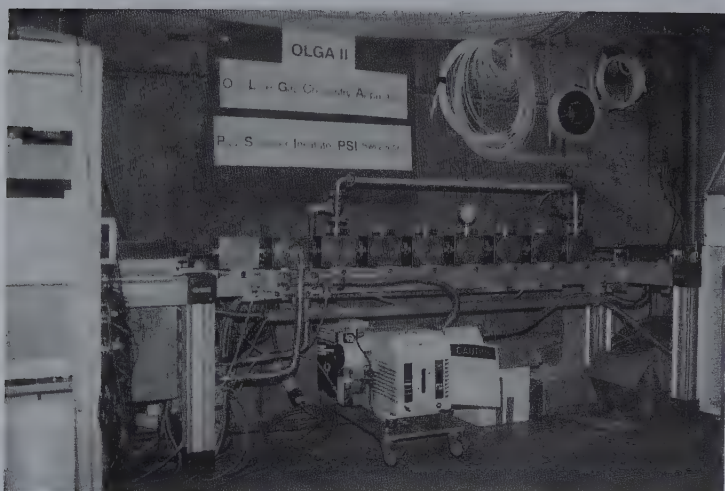


Fig. 9.15. OLGA II, 1990.

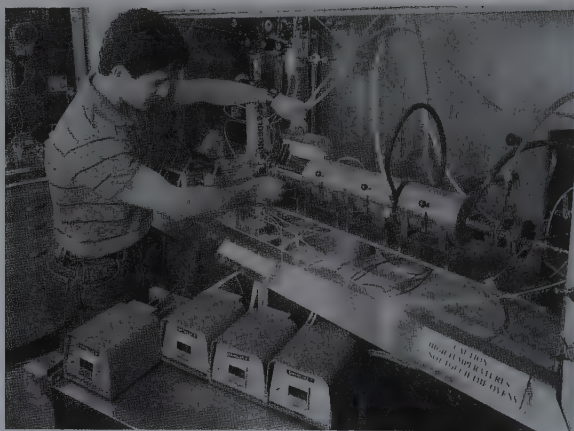


Fig. 9.16. Bobby Kadkhodayan with HEVI, 1992.

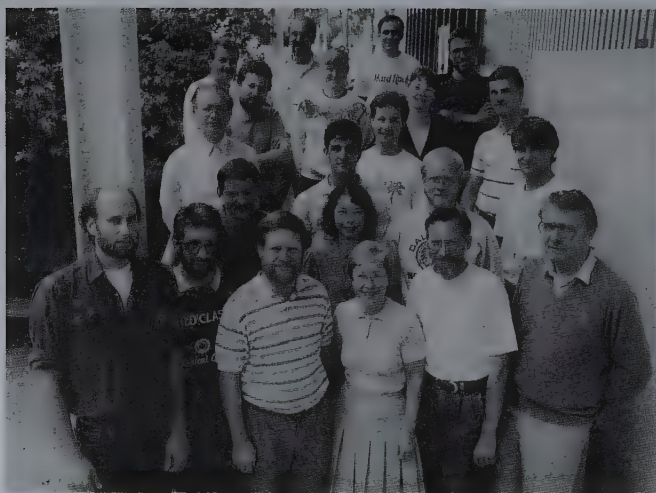


Fig. 9.17. 1990 collaboration to study chemical properties of element 105. *Left to right: bottom row, Ken Gregorich (LBL), Matthias Schädel (GSI), Heinz Gäggeler (Switzerland), Darleane Hoffman (LBL), Jens Kratz (Mainz), Ivo Zvara (USSR); second row, Dieter Jost (Switzerland), Diana Lee (LBL), Ulrich Scherer (Germany), Andy Türlér (Switzerland); third row, Willy Bröchle (GSI), Bobby Kadkhodayan (LBL), Steve Kreek (LBL), Roger Henderson (LBL); fourth row, Peter Zimmermann (Germany), Nancy (Hannink) Stoyer (LBL), Monika Gober (Germany); top row, Erwin Schimpf (Switzerland), Joseph Kovacs (Switzerland), John Leyba (LBL), Alfred Weber (Switzerland).*

Using ARCA, it was shown that hahnium eluted from cation exchange columns with 0.05 M α -hydroxyisobutyric acid (α -HIB), as did niobium, tantalum, and protactinium, but in elutions from triisooctylamine (TIOA) columns with HCl/dilute HF mixtures, hahnium again exhibited non-tantalum-like behavior, indicating formation of oxygen-containing halide complexes similar to those of protactinium and niobium and different from $[\text{TaX}_6]^-$. These results showing the extractability of $\text{Ta} > \text{Nb} > \text{Ha} > \text{Pa}$ were consistent with calculations of complex formation, but predictions of extractability using the Born theory gave the reverse order to that observed. The discrepancy was attributed by Valeria Pershina^{9,43} to multiply charged F^- or mixed $\text{F}-\text{Cl}$ complexes and she suggested that new experiments should be performed with single halide systems for easier comparison with theoretical predictions^{9,44} based on considerations of hydrolysis vs. complex formation derived from electronic structures calculated with an improved Dirac-Slater discrete variational (DSDV) code. These calculations indicated that the extraction order should be $\text{Pa} > \text{Nb} \sim \text{Ha} > \text{Ta}$. First experiments to test these predictions were conducted by a Mainz/GSI/Berkeley collaboration at LBNL in early 1997. Preliminary results^{9,45} performed with ARCA of sorption on Aliquat 336 chromatographic columns from 1 M to 12 M HCl and dilute HF solutions confirm these predictions.

The gas phase studies with both OLGA and HEVI have shown marked deviations in the behavior of rutherfordium and hahnium from predictions based on simple extrapolations of the properties of their lighter group 4 and 5 homologs. The adsorption enthalpies on SiO_2 calculated from a Monte Carlo fit to the measurements of relative yields as a function of temperature between about 100°C and 600°C for the tetrachlorides and tetrabromides of rutherfordium, hafnium, and zirconium show a dramatic break in the sequence for both chlorides and bromides: hafnium is the least volatile while zirconium and rutherfordium are nearly the same (Fig. 9.18). This is presumably due to relativistic effects. The bromides are less volatile than the chlorides, as expected. Data^{9,46} for the group 5 bromides show very low volatility for hahnium relative to niobium and

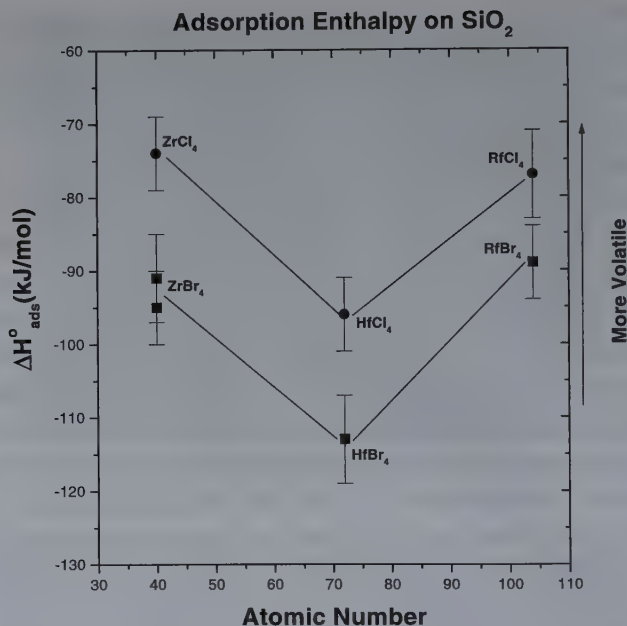


Fig. 9.18. Adsorption enthalpies for zirconium, hafnium, and rutherfordium tetrachlorides and tetrabromides.

tantalum, in disagreement with relativistic molecular calculations,^{9,47} which predict that HaBr_5 should have the highest covalency and lowest effective charge of the group 5 bromides, which should make it the most volatile. It may be that the less volatile oxybromide was actually being formed and additional experiments on the oxybromide are needed.

Hoffman's group at LBNL has performed manual studies to compare the extraction behavior of ^{261}Rf with its lighter homologs zirconium and hafnium and the pseudohomologs Th(IV) and Pu(IV) into tributylphosphate (TBP), TIOA, and TTA from aqueous solutions. These experiments show^{9,48, 9,49} that rutherfordium generally behaves as a group 4 element. However, in extractions from HCl solutions into TBP which extracts neutral complexes, it is found that at low $[\text{Cl}^-]$ concentrations rutherfordium extracts well like zirconium and

hafnium, but at $[Cl^-] > 10$ M rutherfordium extraction decreases rapidly, similarly to Pu(IV). Presumably, this is because rutherfordium and Pu(IV) form stronger anionic chloride complexes at high $[Cl^-]$ concentrations than do zirconium, hafnium, and thorium.

A microcentrifuge system, SISAK III, for continuous separation of nuclides with half-lives as short as a second in combination with a flowing liquid scintillation system, LISSY, for continuous on-line α -spectroscopy, has been developed^{9,50} by G. Skarnemark and his group in Sweden, Jorolf Alstad and his group in Norway, N. Trautmann and his group in Germany, and the Hoffman–Gregorich group at Berkeley. The Hoffman–Gregorich group hosted this collaboration at Berkeley in 1995 and 1996 for preliminary studies of hahnium using 1.8-s ^{261}Ha . In 1996, the behavior of hahnium was also compared with those of its pseudohomolog, Pa(V), using 0.85-s ^{224}Pa . It was possible to detect the ^{224}Pa ^{9,51} and measure its half-life more precisely, but the yield and sensitivity of SISAK still need to be increased in order to provide statistically definitive measurements of the chemical behavior of hahnium.^{9,52}

Hoffman says, "Thus our group was instrumental in initiating what has turned out to be a long and very fruitful series of international collaborations to study the chemical properties of the heaviest elements and the influence of relativistic effects. These collaborations, involving the instruments ARCA, OLGA, HEVI, and SISAK-LISSY, were hosted primarily at Berkeley and later at GSI. The aqueous chemistry of hahnium has been studied in a variety of systems and the volatilities of the halides of both rutherfordium and hahnium have been examined. Together with our group's manual studies of the aqueous chemistry of rutherfordium, we have shown that the aqueous and gas phase properties of rutherfordium and hahnium cannot be reliably extrapolated from those of their lighter homologs in groups 4 and 5 of the periodic table. In some instances rutherfordium even behaves more like its pseudohomologs, Pu(IV) and Th(IV), and hahnium sometimes behaves more like its pseudohomolog, Pa(V). This concentrated experimental effort has provided much data and the impetus for theoretical chemists to

compare their predictions of relativistic effects on chemical properties with the experimental data and to refine and develop their predictive models."

There are several recent reviews^{9.53–9.55} of the chemistry of the transactinides which compare experimental results for elements 104 and 105 with theoretical predictions and discuss the prospects for extending the studies to still heavier elements. Valeria Pershina (see photo in Fig. 9.19) has given an excellent review^{9.56} of the theoretical studies of transactinide compounds as of early 1996, and of recent^{9.57} progress that has been made in predicting chemical properties. She herself has pioneered in the prediction of the extraction order for the group 5 elements in pure halide acid solutions based on considerations of hydrolysis vs. complex formation from electronic structures calculated with an improved Dirac–Slater discrete variational (DSDV) method. The preliminary experiments performed at the Berkeley 88-Inch Cyclotron in 1997 by the Mainz–GSI–LBNL collaboration seem to confirm her predictions. Thus careful design of



Fig. 9.19. Valeria Pershina together with (left to right) Matthias Schädel, Sigurd Hofmann, Heinz Gäggeler, Jens Kratz, and Jorolf Alstad at the Banquet of the 41st Welch Conference on Transactinide Elements, Houston, Texas, October 1997.

experiments so that they can be directly compared with theoretical predictions is proving especially valuable in our attempts to gain a more complete understanding of the underlying phenomena involved.

The results for rutherfordium and hahnium have provided the impetus for the formation of an expanded collaboration including scientists from Dresden/Rossendorf, the former Soviet Union, and Japan to study the properties of seaborgium at GSI in 1995, 1996, and 1997. (See Chapter 10.)

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Chapter 10

Seaborgium

10.1. Discovery

The element with the atomic number 106 was synthesized and identified in 1974 but was not given a name until 1994. The investigators^{10.1} who have been officially sanctioned by the IUPAP/IUPAC Transfermium Working Group (see Chapter 13) as the discovery team were a group from the Lawrence Berkeley Laboratory (LBL) — Albert Ghiorso, J. Michael Nitschke, Jose R. Alonso, Carol T. Alonso, Matti Nurmia, and Seaborg — and from the Lawrence Livermore National Laboratory (LLNL) — E. Kenneth Hulet and Ronald W. Loughheed (Fig. 10.1). The experiment was performed at LBL's Heavy Ion Linear Accelerator (HILAC) by bombarding ^{249}Cf with ^{18}O to produce the isotope $^{263}106$. The new nuclei were shown to decay by the emission of alpha particles with a half-life of 0.9 s and a principal alpha energy of 9.06 MeV to the previously known $^{259}_{104}\text{Rf}$, which in turn was shown to decay to the known $^{255}_{102}\text{No}$. Thus the atomic number of the new nucleus was firmly established by a genetic relationship to its daughter and granddaughter.

At about the same time another claim^{10.2} to the discovery of element 106 was made by a Russian group working at the Laboratory of Nuclear Reactions at the Joint Institute for Nuclear Reactions in Dubna, Russia: Yuri Ts. Oganessian, Yu. P. Tretyakov, A.S. Iljinov, A.G. Demin, A.A. Pleve, S.P. Tret'yakova, V.M. Plotko, M.P. Ivanov, N.A. Danilov, Yu. S. Korotkin, and G.N. Flerov. They reported the observation of a spontaneous fission activity with a half-life of 4–10 ms, produced by bombarding ^{208}Pb with $^{54}_{24}\text{Cr}$, which they



Fig. 10.1. The codiscoverers of element 106, seaborgium (Sg). *Sitting, left to right:* Matti Nurmia, E. Kenneth Hulet, Glenn T. Seaborg, Albert Ghiorso, and Carol T. Alonso. *Standing, left to right:* Ronald W. Loughheed, J. Michael Nitschke, and Jose R. Alonso.

assigned to $^{259}_{106}$ on the basis of reaction systematics. Ghiorso recalls that "the Dubna disclosure came dramatically in a private meeting after we had told him of the production and identification of our isotope, $^{263}_{106}$. Both sides were completely surprised and, not wanting a repeat of the element 104 and 105 controversies, were quick to say that they did not want to get into another public argument as to which side had found the element first. The immediate consequence was that it was agreed on the spot that the discoveries were essentially simultaneous and neither side would claim the right of discovery until it had been determined which of the findings would be confirmed by subsequent work. It was agreed that if the discoveries of both groups were proved to be correct, then they would name the new element jointly. Because of the competing claims, the two groups agreed not to propose a name for element 106

until it could be determined which group had priority for the discovery.

In 1984, Demin *et al.*^{10.3} at the Dubna laboratory showed that the SF activity attributed in 1974 to element 106 by the Flerov group was actually due primarily to the SF from the ²⁵⁶104 daughter of element 106, and not to element 106 itself, thus effectively invalidating the Dubna claim to the discovery of element 106.

The following is the “untold story” of the earlier production of element 106 in 1970, which could have resulted in the actual discovery of the element if the data had been carefully analyzed and the experiments repeated at that time. The story is in Al Ghiorso’s own words.

10.2. The “Untold Story” of Seaborgium

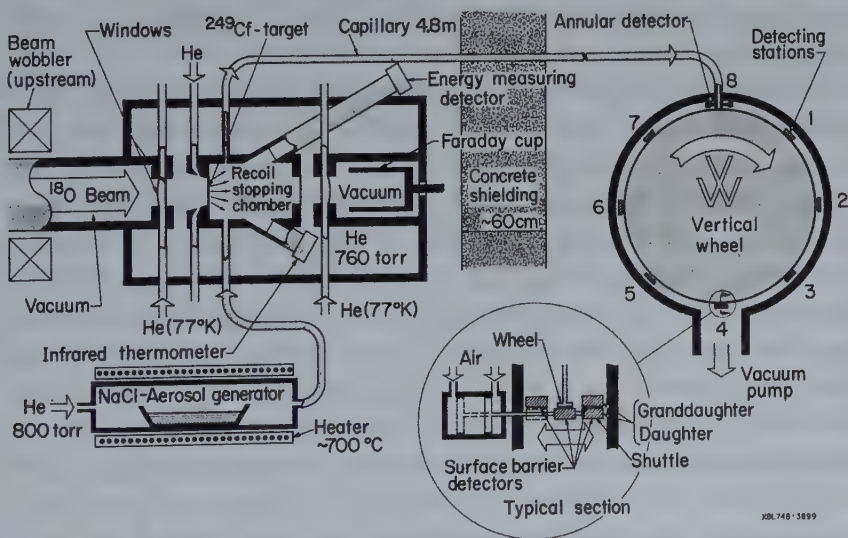
“The story of the discovery of element 106 spans more than two decades. The element was actually first observed in 1971, three years before the definitive experiments which allowed the discoverers to announce that it had been found. If the circumstances of that first finding had been only slightly different the element 106 discovery would have been made with certainty at that early date and, since there was no competing claim at that time we would have given a name to the element. What that name would have been is unknowable except that it would not have been ‘seaborgium’! Fortunately, two decades of history crammed with the discovery of six more elements intervened to make the difference. This is the story of that element.

“The first effort to produce an isotope of element 106 followed shortly after the discovery of element 105, hahnium, in 1970. The genetically based vertical wheel (VW) apparatus, which had been so effective in discovering elements 104 and 105, had undergone a continuous process of improvement [Fig. 10.2], with gratifying results. The special target of ²⁴⁹Cf had done yeoman’s service in demonstrating its ability to produce α -particle emitters which could be identified as coming from elements 104 and 105 when it was bombarded by carbon and nitrogen ions from the HILAC. In these

(a)



(b)



XOL 740-3899

Fig. 10.2. (a) Improved vertical wheel (VW) used in the discovery of seaborgium, 1974.
 (b) Schematic of improved VW showing all seven detecting stations.

cases, many events had been observed and there was no uncertainty about the interpretation of the results. As a final experiment on the old HILAC, ^{18}O ions were also tried on this target just in case it might be possible to also see element 106, and in a period of a day three candidate events at an α -energy of about 9.1 MeV were observed. This energy qualified as a value that we expected to see, but the background was not completely zero and we had no confidence that the events were meaningful, so we did not publish this work. Nor did we follow it up soon thereafter, because the HILAC was shut down immediately after this experiment to convert it into the SuperHILAC. The conversion took much longer than we had expected, and when it did come back on, other new experiments looking for superheavy elements took precedence in our scheme of things. What we did not think of at the time was that there was another way of analyzing the data that would discriminate against the small background that was present and that data were on computer tape. If we had made this analysis during the shutdown, new data would have been our top priority and we would have confirmed the 1970 work as soon as the SuperHILAC began to operate a year later.

"The SuperHILAC evolved in a very unusual way. In 1964 Bob Main, Bob Smith, and I had invented the Omnitron, a bold new concept for a two-ring synchrotron system for both acceleration and storage that would accelerate ions of *all* of the elements within an energy span that went from zero to the GeV range. We developed the concept because I had been persuaded by Cornelius Tobias and John Lawrence that a high energy heavy ion accelerator would be a real breakthrough for medical science in the treatment of various types of cancer. We knew that it would also be an excellent accelerator for both low and high energy physics. By a fortunate coincidence, the idea of the Magic Island of Superheavy Elements had just been broached by Bill Myers and Wladek Swiatecki in great detail. It immediately became clear to us that our proposed machine would be very attractive for making a comprehensive search for these "superheavy elements" by allowing the experimenters a wide choice

of bombarding ions. This was very exciting, because now the Omnitron could be shown to encompass a wide range of sciences and thus garner more support.

"Seaborg happened to be the Chairman of the Atomic Energy Commission at this time and because of his continuing interest in the heavy element end of the periodic table it was not difficult for me to convince him that the Omnitron was an excellent idea. Funds were made available to allow our very competent team of designers and engineers to make a vigorous study. This was done during the next couple of years and ended with a proposal to build our dream machine. It had an estimated cost of about \$24 million (1971), and since the AEC was very much in favor of it, design and construction plans were launched to build the Omnitron at Berkeley. Most unfortunately, it had to compete with the political forces in the US who were pushing the disastrous Vietnam war. (It is worth noting that the US was spending on the war machine the equivalent of at least three Omnitrons each and every day at that time!) Being aware of the possibility that our new accelerator might not be funded we had also developed a 'fallback' position, the improvement of the old HILAC into the SuperHILAC. It did not have the high energy capability nor was it as versatile at low energies as the Omnitron, but it was a variable energy high intensity accelerator and consequently a vast improvement over the original HILAC.

"It turned out that there was another fatal impediment to the construction of the Omnitron and that was the fact that the famous 200-BeV proton accelerator was turned down at this time for construction at Berkeley by the AEC as the next big high energy facility. Instead, after careful studies had been made by national review committees appointed by the AEC it was decided in 1967 to put the \$250 million machine in Illinois, a site that was to become Fermilab. Without going into the technical reasons why Berkeley's 200-BeV design was rejected, suffice it to say that Seaborg was the Chairman of the AEC at that time and so naturally he was blamed by some in the Berkeley Physics Division for this crushing blow, and consequently they were in no mood to be appeased by that upstart

machine of the nuclear chemists called the Omnitron as a consolation prize.

"Without the backing of the Laboratory Director expensive projects have almost no chance of getting funded, especially in an overburdened AEC budget. Twice before Seaborg had intervened by going all the way to President Johnson to keep the Omnitron in the budget. This problem was further compounded by the fact that the \$100-million Stanford Linear Accelerator Center (SLAC) had just been completed and needed more funds for operation. The Stanford people, *with the acquiescence of McMillan*, arranged a deal with the JCAE to transfer to SLAC the first \$2 million that had been promised to Berkeley for the Omnitron. The promise was made that the \$2 million would be put back into the budget for the Omnitron the following year. The importance of this money was that it would have funded the necessary preliminary work called A&E and, traditionally, if a project got this far its chances of going all the way were extremely good.

"When Seaborg asked me the basic question as to whether we wanted to try for the Omnitron again the next year and risk losing it once more or switch over to our fallback position, the SuperHILAC, I decided to go for the latter. At the time this seemed to be the only choice that we had, since it was ten times less expensive to upgrade the HILAC than it was to build the Omnitron. The driving incentive was that the superheavy elements at that time seemed tantalizingly close and we needed a better accelerator now to look for them. We did not want to wait for years!

"This is how the SuperHILAC came to be constructed at Berkeley. It would prove to be very useful for the next 20 years, but there is no question that the Omnitron, 25 years ahead of its time, would have been a much better accelerator for Berkeley and the world. It would have changed the history of the Laboratory in very significant ways.

"The metamorphosis of the old HILAC into the new SuperHILAC took a remarkably short time, only some 15 months, thanks to the genius of Bob Main. The shutdown generated a plaque, shown in



Fig. 10.3. Plaque to commemorate the demise of the old HILAC on Feb. 5, 1971. *Left to right:* Bob Main, Bill Stahl, Frank Grobelch, and Al Ghiorso.

Fig. 10.3, to commemorate the event. The new accelerator, for that is what it was, consisted of two new linacs with brand-new, innovative technology and a new injector that had been built as part of the Omnitron development program. The contributions from the old HILAC were the building, the original low voltage injector, and its infrastructure. The SuperHILAC was ready for initial use in 1971 but, as might have been expected, had a lot of teething problems, partly because of the new technology and partly because it had also become the injector for the BevaLAC.

"Although our initial experiments were devoted to simple searches for the superheavy elements, we were well aware of the likelihood

that we should also be able to produce element 106 by bombarding the reactor-produced isotope ^{249}Cf with ^{18}O ions. Some years before, as a member of the National Transplutonium Program Committee, I had suggested that the nation's total supply of berkelium, which β -decays with a nine-month half-life, be set aside to grow this isotope for experimenters who wanted to use monoisotopic californium. The Committee had agreed and a policy had been established to try to optimize the production of ^{249}Cf . This is the material that enabled the Berkeley team to discover elements 104 and 105, and now it was going to produce element 106.

"The VW detection system which had been so productive in discovering elements 104 and 105 was gradually improved in many ways for use with the new SuperHILAC. (1) Mike Nitschke replaced edge-cooling, the usual technique at that time for cooling targets, with a gas-cooling method and gained a factor of 2 or 3 in beam intensity that could be safely handled. (2) To make a more durable target to withstand the larger beam current Hulet and Loughheed developed the techniques for vaporizing californium trichloride onto a thin titanium substrate to replace the traditional electrolytic method. This process also efficiently removed lead from the target material; this element, even as a minuscule contaminant, was the bane of many of the experiments with actinide targets because it produced high energy α -emitters that mimicked the transplutonium activities. (3) The VW itself was moved five meters away into its own cave to reduce neutron radiation of the detecting crystals. This meant that the recoil atoms would have to be transported in a fraction of a second by an aerosol (a suspension of fine particles of NaCl) inside a capillary tube by flowing helium gas that was jetted onto the periphery of the wheel. Mike Nitschke and I devoted many months to this problem before we came up with a simple system that would also be highly efficient. Another important addition was the incorporation of a new type of computer analysis wherein we could follow the decay by α -particle emission from element 106 through element 104 by a time correlation of two successive α -particle decays

that emanated from the same spot where the atoms of element 106 were collected. This genetic analysis was a powerful tool and supplemented the physical correlation that we had observed by recoil transfer in the cases of elements 104 and then 105.

"Finally, in June of 1974, we were ready to try again. It occurred to me that it would be interesting to test out our new computer program by reanalyzing the data taken in 1971, so I suggested to Jose Alonso that he re-examine the three interesting events that we had seen at that time before we started our new runs. Much to the joyful amazement of everyone, on 8 June he found that the three events which we had singled out showed an α - α time correlation of a couple of seconds and the other ancillary data agreed with the interpretation that they were probably due to $^{263}\text{106}$ decaying to $^{259}\text{104}$. In the Log Book for that day I wrote down this comment: 'Wow! Do you mean that we found element 106 on 24 Jan. 1971 and didn't report it!'

"That was on a Saturday when we were making our final checks. Our beam time would begin two days later, when I had scheduled five days and nights around the clock for the big experiment. There were many groups from all over the country who also were anxious to use the new accelerator in their own premier experiments, so beam time was at a premium. I was the one who scheduled the beam time and I had to be scrupulously fair in allocating it. It was evident from the schedule that our group had not had an inordinate amount of time for testing out the new system but we felt confident that the equipment was ready to go. Because Jose had found the three old correlations we seemed to have a guaranteed result, so we were anxious to start our runs and see if we could repeat that work.

"Ken Hulet and Ron Loughheed had vaporized a beautiful ^{249}Cf target onto a thin titanium backing for the new experiments and after the usual preliminary checks the run with ^{18}O ions proceeded. Before long we began to see α -particle events very similar to the old ones. This time, because of the improvements, we had a higher counting rate and a lower background and we slowly continued to accumulate

the data that we would need to prove that we had observed a new element. As before, the new nuclei were shown to decay by the emission of α -particles with a half-life of 0.9 s and a principal α -energy of 9.06 MeV to previously known ^{259}Rf (8.77, 8.86 MeV) with a time correlation between the two alphas in the range of 2–3 s. The latter nuclide in turn was shown to decay to the known 3-min ^{255}No . The atomic number of the new parent nucleus was thus firmly established by the genetic relationship to its daughter and granddaughter. Now there was no question. We had found element 106!

“But the Dubna scientists had been busy also. This same year (1974) had marked the discovery by Oganessian and coworkers of the “cold fusion” reactions to make heavy elements. After successful tests in which his group showed that fermium isotopes could be produced with nanobarn cross sections by bombarding ^{208}Pb with ^{40}Ar , he decided to make use of the same principle to produce an isotope of element 106. He began to bombard ^{208}Pb with the rare isotope ^{54}Cr , using their rotating wheel system equipped with SF track detectors as the detection tool (see description in Chapter 9). In experiments performed two months before our June runs, they had succeeded in finding small but finite amounts of SF activity which they thought could be coming from element 106. In these first experiments they thought that there were two SF activities with half-lives of 1 and 10 ms due to $^{257}\text{106}$ and $^{259}\text{106}$, respectively. It so happened that the International Conference on Reactions in Complex Nuclei was scheduled to be held June 10–14, 1974, in Nashville, Tennessee, so they let it be known that they would like to present a paper on their recent work. They did not divulge to the Conference exactly what they were going to talk about but they let it be known that it had something to do with the heavy elements and that G.N. Flerov, himself, would attend. This would be his first visit to the US and rumors immediately began to emerge from Oak Ridge that they had found element 106. When it became known that Flerov would be in the country Seaborg invited him to visit LBL also and it was arranged that he would come to Berkeley after the Nashville Conference.

"Originally, Nitschke and both Carol and Jose Alonso had been scheduled to go the Conference, but an unforeseen opportunity had arisen for SuperHILAC beam time and I decided to take advantage of it for an element 106 run. Only Carol went to Tennessee to present a nuclear hydrodynamics paper that she had been working on and to keep an eye on what happened at the meeting with regard to element 106. Each day she reported to me what was going on and as a result I got a wonderful blow-by-blow description of what happened in Nashville.

"At this time, with the Cold War still in progress, there was, inevitably, an intense rivalry between Dubna and Berkeley with regard to which laboratory would garner the prestige that would naturally be bestowed on the first group to discover element 106. The Dubna group had been bloodied by the encounters on elements 104 and 105 with our laboratory and they were intent on salvaging their reputation by being first with element 106. Carol was aware of this tension and was not certain what to do if the Russians announced that they had indeed found the element. Should she immediately announce our results to counteract theirs? At that time it seemed important to tell the world immediately what the situation was, so Carol called me at Berkeley to find out the latest in the ongoing experiment and to ask whether it would be proper for her to announce our discovery.

"The big question in our minds was whether Flerov would announce anything at all about their attempts to find the element. Many in the audience were SuperHILAC users, so they knew something about heavy elements research and its colorful history; by rumor and direct contact, they soon became aware of this playlet that was being enacted before their eyes. They too began watching every move of the Russian delegation. Neither side knew what the other's cards were and whether there were any trumps or wild cards floating around.

"It turned out that there *was* a wild card — Carol soon found out that Curt Bemis and his group at Oak Ridge were going to start doing the same experiment as we were the following week! There was one

important difference, however; their detection system, though very powerful, was inherently very inefficient, being centered on proving the Z identification of element 106 by measuring the characteristic X-rays of the element 104 daughter produced by α -decay of element 106. This technique has one important advantage in that it is an absolute one and, given enough events, can identify a new element with certainty. It had been first demonstrated a few years before in the case of element 104 when Bemis *et al.*^{9.23, 9.32} had brilliantly confirmed the Berkeley discovery of $^{257}\text{104}$ and later 105. This time they intended if possible to discover element 106 at least simultaneously with our work, if not before. Bemis had heard a rumor that we had found $^{263}\text{106}$, so he was interested in knowing the half-life of the isotope so that he could set up his experiment most efficiently. Any information that they could get about the ongoing Berkeley experiment would be helpful to them. I told Carol that there was at least an order of magnitude difference in sensitivity between their method and ours and that I did not think that they could do it. [Note: The Oak Ridge experiment was successful in observing a total of 30 α -particles of $^{263}\text{106}$ but, unfortunately, not a single X-ray in coincidence with an α -particle. Thus, though they observed the same α -energies (9.06 and 9.25 MeV) and half-life (0.74 ± 0.23 s) as Berkeley, they did not have any Z identification of the activity, so their work could not be considered a confirmation of ours.]

"So Carol found herself in the middle of all this — Flerov about to say something, the Oak Ridge people aware that we had some data that would be useful to them, and she herself wondering if she should say something about our work if Flerov announced that they had found element 106. The game now was to find out what Flerov was going to say. An opportunity arose on the second day of the Conference at a 'posh' party for the speakers that was held on a paddle wheel boat making its way slowly down the Cumberland River. Carol was immediately taken in tow by Curt Bemis, who wanted to know more details of the Berkeley experiment. She was not sure how much she should divulge, so she merely said that we had been successful and that he should call me in Berkeley for more

information since everything was very preliminary. Bemis felt that we should announce our discovery if Flerov announced theirs, and volunteered to find out what their plans were. Carol, the only woman on the boat, ensconced herself at the end of the boat near the paddle wheel downstream from the others and there various people in turn reported to her what they had found out. Allan Bromley, Jack Miller, Bill Myers, and Curt Bemis all went to the Russians in turn and asked them in subtle ways if they were going to announce element 106. At one point Flerov answered, 'No, 108!', but he was only having his little joke. By the time the boat got back to Nashville the whole boat was alive with whispers and rumors.

"Exhausted from the party, Carol called me late in the evening and reported what had happened. I told her that we had run into various problems, that things had gone badly, probably because we had been up for two days running and were not operating efficiently, and that we might have to give up on this run. We had enough data to announce but I was holding out for one more run before we were ready to publish.

"Our normal course of action was to issue a press release on our discoveries only when a paper had been sent off for publication. I told her that although we continued to find α - α correlations, I felt that there were a few places where we could improve our insight into the data and it would be better to wait. I told her that I thought it would be better to let the Russians go out on a limb and just watch to see if it got chopped off. Of course, Carol was disappointed, but she kept silent. Seaborg happened to be in Sweden attending a superheavy elements conference at this same time and I kept him informed by phone of the drama that was taking place in Tennessee. I found that he agreed with me about withholding any announcement at that Conference.

"The next day, Wednesday, was sort of rambling until Bemis turned up with a copy of Flerov's paper to show to Carol. The organizers of the Conference had insisted that they should have a copy for the Conference report as a condition for it to be presented. It was to be delivered on Thursday and it had just one sentence in

it about finding element 106 in a Cr-plus-Pb experiment. There were no details at all! This raised eyebrows and also suspicions that there must be something marginal about their experiment. Carol reached me in Berkeley with the news and I indicated that we should still maintain our cool strategy and not be panicked into doing anything premature. I cheered her up by saying that we had overcome our difficulties, we would continue to see good 106–104 correlations, and as a consequence, Jose would be arriving at Nashville the next day. I said that I had pre-empted more beam time so that we could run for a few more days and that we would even be running during Flerov's visit to Berkeley the following week.

"On Thursday afternoon Ivo Zvara got up to deliver Flerov's talk in English to save the usual delays when a speech in Russian had to be translated into English. He reported on a number of SF activities, including new light isotopes of element 104. When he got to the middle he read the sentence about element 106 and there was no reaction at all from this sophisticated audience. At the end of the talk, Bromley looked over at Carol and Jose to see if they were going to comment by mentioning the ongoing Berkeley work. Carol slowly shook her head. It had been agreed by the Alonsos, Bemis *et al.* that the best plan would be for everyone to ignore the Dubna claim, and a tentative press conference was thereby canceled. After the break following the Flerov talk, Carol made an excellent impression on the audience when she gave her talk complete with movies about nuclear hydrodynamics. This had nothing to do with the element 106 work, but it did show that she was in the forefront of another complicated field.

"On the following Monday, Flerov arrived in Berkeley with his entourage [Fig. 10.4], which included V.A. Karnaukhov, V.V. Volkov, Ivo Zvara, and K.-H. Kaun. After the usual greetings they were taken up to the SuperHILAC to see the element 106 experiment in action. They were really surprised to see the sophisticated nature of our experiment and how we were really proving that 'one alpha is worth a thousand fissions,' the statement that I had been taunting them with for the preceding decade. Though the excellent data that



Fig. 10.4. G.N. Flerov briefing at SuperHILAC, June 17, 1974. *Left to right:* R. Otto, E.A. Norris, I. Binder, T. Raunemaa, R. Main, A. Ghiorso, J.M. Nitschke, G.N. Flerov, J.O. Rasmussen, G.T. Seaborg, V.V. Volkov, E.K. Hulet, I. Zvara, V.A. Karnaukhov, R.M. Diamond, K.-H. Kaun, and J. Kratz.

we had accumulated so far had already proved that we had observed $^{263}_{106}$, we indicated that we intended to run a little longer before we published our work. We could tell that Flerov and his group were surprised at how well our experiment had gone.

"Flerov's response was to tell us about their Pb-plus-Cr experiments, wherein they had managed to detect a small number of SF events which they thought were coming from $^{257}_{106}$ and $^{259}_{106}$ with half-lives of about 1 and 10 ms, respectively. At that point we had no way of judging the validity of their results, so we had to assume that they could be correct. As a result there was mutual agreement that both sides should forego naming element 106 until the work had been confirmed in some way, to forestall the kind of public conflict that had arisen with elements 104 and 105. If the work of both laboratories was found to be correct we agreed that we would name the element jointly, since the discoveries were essentially simulta-

neous. Note that we did not address the question as to whether the Dubna experiments had actually proved that the SF activities were indeed due to element 106. The identification consisted entirely of the fact that they had bombarded element 82 with element 24 at an energy near the Coulomb barrier and found SF activities.

"We continued to make more bombardments on and off for three more months to be absolutely certain of the identification of $^{263}106$. For this purpose new californium targets had to be made to overcome various problems as they arose — and altogether Ron Loughheed and Ken Hulet made four targets of ^{249}Cf ! This was a tremendous undertaking that involved a lot of chemistry and careful attention to details.

"We celebrated with one of the famous 'HILAC Punch' parties, to which many Berkeley researchers were invited. All the 106 discoverers were given specially marked 106 hats and paperweights, and a good deal of speechifying and merrymaking ensued, including a number of joke presentations and a wall size SuperHILAC board game complete with heavy element 'dragons and pitfalls.'

"As recounted earlier, the Berkeley work was duly published in 1974 in *Physical Review Letters*^{10.1} by a team which consisted of Ghiorso, J. Michael Nitschke, Jose R. Alonso, Carol T. Alonso, Matti Nurmi, and Seaborg from the Lawrence Berkeley Laboratory (LBL), and E. Kenneth Hulet and Ronald W. Loughheed from the Lawrence Livermore National Laboratory (LLNL), and the Dubna work was published the same year by Oganessian *et al.*^{10.2}

"After publication in 1984^{10.3} of the very carefully done new experiment by a different group of Soviet scientists headed by A.G. Demin, one of the original group that observed the SF emissions, showed that the short-lived SF activity that had been seen at that time was almost entirely due to $^{256}104$; it was now quite clear that the Dubna scientists had not observed element 106 in 1974 after all — they had measured its daughter produced by α -decay of $^{260}106$!"

10.3. Independent Confirmation and Naming of Element 106

Although the Tranfermium Working Group had credited the discovery of element 106 to the Berkeley–Livermore group in 1992, the group did not immediately propose a name. This was because in 1976 an international group of scientists had proposed more stringent criteria^{10.4} for the discovery of new chemical elements. Included among these was the suggestion that a name should not be proposed by the discoverers until the observation had been



Fig. 10.5. 1993 photo of the Hoffman Research Group together with three of the original discoverers of seaborgium. *Front row, left to right:* Al Ghiorso,[†] Ken Gregorich,^{*} Mike Mohar,^{*} Yas Watanabe (Visiting Scientist from Japan), Glenn Seaborg,[†] Darleane Hoffman,^{*} Diana Lee,^{*} Chris Kacher,^{*} Mike Lane,^{*} Tonya Pouncey (Administrative Assistant). *Back row, left to right:* Nancy Stoyer, Jerry Forinash (undergraduate), Dawn Keeney, Jerry Yang (undergraduate), Erik Osterreider, Alan Veeck, Mary Neu, Eric Sylwester,^{*} Matti Nurmia.[†] (^{*}Members of the Group involved in the 1993 confirmation experiment. [†]Members of the original discovery team.)

confirmed independently by another group. In accordance with this suggestion, a different LBL group (Fig. 10.5) decided it was important to confirm the original discovery in order to clear the way for the LBL/LLNL discovery group to propose a name for element 106. In 1993, the LBL group, led by Ken Gregorich and Darleane Hoffman, finally obtained approval from the Program Advisory Committee of the LBL 88-Inch Cyclotron for their proposal to attempt to confirm the earlier discovery. They produced $^{263}\text{106}$, via the identical $^{249}\text{Cf}(^{18}\text{O},\text{4n})^{10.5}$ used in the discovery experiment of Ghiorso *et al.*^{10.1} However, they used the 88-Inch Cyclotron rather than the HILAC and their different horizontal rotating wheel system in a special parent–daughter mode to positively identify $^{263}\text{106}$ by correlating its decay with the decay of its known 3.1-s ^{259}Rf daughter. A half-life value consistent with that of 0.9 s and a cross section consistent with that of 0.3 nb reported by Ghiorso *et al.* in 1974 were obtained, thus completely confirming the original discovery.

At about this time pertinent experiments at GSI also showed that $^{259}\text{106}$ was not an SF emitter and $^{260}\text{106}$ was predominantly an α -emitter so that neither isotope had the characteristics observed by the Dubna group.

The Berkeley–Livermore group then began serious consideration of names for element 106, since not only had their discovery been credited by IUPAC, but it had now been confirmed by an independent group as well.

Then Ghiorso took the initiative and suggested another approach. Al Ghiorso's first hand account of this unique naming process follows:

"Now we felt that our claim to the discovery of element 106 stood alone and we should think about christening it. The problem was that there were many possible names, since each of the codiscoverers had a favorite. My personal choice was to call it 'alvarezium,' after the great Luis Alvarez. He was one of the world's great physicists and had been responsible for many fundamental developments and discoveries that had been crucial in advancing nuclear physics. He was also the one who had made our experiments possible by

inventing the modern linear accelerator which bears his name. In particular, he had suggested that a heavy ion linear accelerator should be built for our heavy element research program and that it should incorporate the new idea of magnetic strong focussing inside the drift tubes. But my seven colleagues did not seem to have the same awe of Luis that I had, so they were not persuaded by my suggestion and came up with their own ideas.

"Some of these suggestions for honorees were: the eminent French nuclear scientist Frederic Joliot (a transfer of the Dubna suggestion for naming element 102); the scientific giant Sir Isaac Newton; the famous inventor Thomas Edison; the famous scholar and inventor Leonardo da Vinci; early explorers such as Christopher Columbus, Ferdinand Magellan, and the mythical Ulysses; the great American statesman George Washington; a Russian scientist such as Peter Kapitza or Andrei Sakharov; and Finland, the native land of a member of the discovery team. These were all good names, as I was the first to admit, and for a while I despaired that we would ever be able to reach a consensus on this real dilemma.

"The problem was solved in a very unusual way. When the confirmation of our 1974 discovery of $^{263}106$ was published in *Physical Review Letters* in 1994, I received a telephone call from Malcolm Browne of the *New York Times*. We had often talked by telephone about the Berkeley work in the heavy element field and he was very knowledgeable in that regard. He congratulated me on the confirmation and then asked me a leading question 'What are you going to name element 106 — ghorsium?'

"This was an old joke, since I had been associated with the discovery of so many elements. In fact, in 1957 at the Nuclear Chemistry Division Christmas Party, Seaborg had been given a special Christmas present in the form of an enormous box filled with packing material and boxes within boxes like a Russian matrioshka doll, the center one containing a large bottle labeled *Element 110, Ghorsium* (Fig. 10.6). Earl Hyde had written a clever caption containing the following lines:

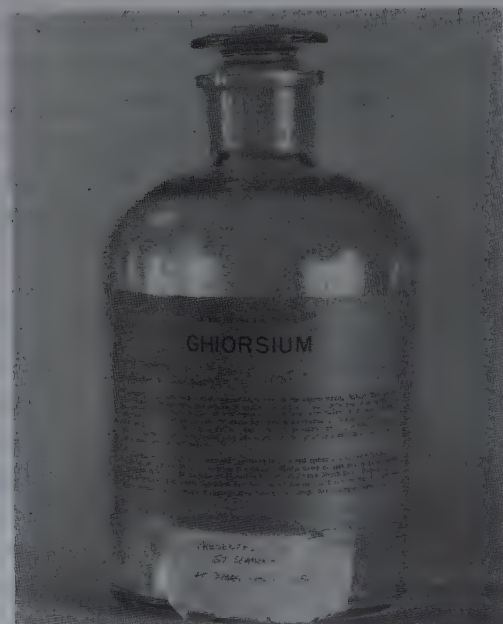


Fig. 10.6. Bottle of ghiorcium.

'A Weightless Sample of ^{299}Gr

GHIORSIUM

Atomic Number: 110

Preparation: Shot from Guns

Properties: A worthless metal named in honor of its discoverer, Albert Ghiorso, can be prepared only between the hours of midnight and 6 a.m. Only one atom of this element has been prepared but it has been conclusively identified as to atomic number and chemical properties by the ion exchange delusion method. Spontaneously inflammable. Decays by alpha decay, beta decay, positron decay, orbital-electron capture, spontaneous fission and just generally falls apart in a hell of a hurry. Also has an automatic transmission.

The nucleus of the new element, ghiorcium, is not spherical or football-shaped but has a shape resembling a summer squash. When news of this reached Copenhagen, Professors Bohr and Mottelson

drowned themselves in the North Sea. Fortunately, it is not necessary to solve the complicated equations of rotational motion for this odd-shaped nucleus as it always disintegrates before it completes a single revolution.'

"I still have the bottle as a fond memento of my longtime colleague. Earl Hyde passed away on March 3, 1997. He was a very important member of the scientific world.

"But back to Malcolm Browne. I laughed at the idea of naming element 106 'ghiorsium' but it got me thinking — why not name it after a living person? In the cases of Einstein and Fermi, whose names we gave to elements 99 and 100, the fact that they were alive when we proposed their names seemed irrelevant. We all felt that they were immortal. But this was different. We knew that there could not be many more elements produced and we should be very careful of our choice.

"As with many ideas in my career, my subconscious worked on the problem for a little while and a few nights later I awakened from a deep sleep with the strong feeling that now was the perfect time to propose the name of a living person. I asked myself the question, Why not name it 'seaborgium'? I felt that in the great panoply of names that had already been picked for the heavy element region, the name 'seaborgium' would be of equal worth to curium, einsteinium, fermium, mendelevium, lawrencium, rutherfordium, hahnium, nielsbohrium, and meitnerium.

"This was a very serious step to take, so I thought that I had better test it out in a number of ways. First I tried out the idea on my wife, Wilma. She had no hesitation in saying that it would be a great name, so I decided to go ahead cautiously and see how my codiscoverers felt about it, one at a time. I happened to run into Jose Alonso first and was pleased to find that he also thought that it would be a great name. He had the same hesitation that I had — namely, would Glenn accept it? We both felt that the overriding consideration should be that we should not do anything that would embarrass Glenn. The next day Jose told me that Carol

also was very enthusiastic and I knew that we were on the right track. Encouraged by this initial reception I went ahead and gradually canvassed the other six members of the team (excluding Glenn, of course).

"Mike Nitschke was not sure at first, since he was involved in an abortive scheme hatched at GSI when he was there a couple of years before. He and Armbruster were playing with names for all of the elements and Glenn's was in one of the permutations. Mike had talked with Glenn and found that he was against it at that time, so he felt that it would not be acceptable now. I pointed out that it was a completely different situation now. He agreed that it was worth a try, so I continued to try to build a consensus.

"Matti Nurmia, who had just returned from Finland, was my next contact and he too agreed that it would be very appropriate and was all for it. At about this time I started to consult a few people outside the group of codiscoverers: Darleane Hoffman, Earl Hyde, and Paul Karol of Carnegie-Mellon come to mind, and I found that there was no hesitation — everyone was enthusiastic. I was surprised to hear from Sam Markowitz (UC Berkeley) in a voice mail message; he had independently come up with the same idea. He wrote up his thoughts and reasons and I received them in the mail a few days later.

"The next person to be contacted was Ron Loughheed. Again there was agreement and now I knew that if Ken Hulet agreed, we had achieved a consensus. Having progressed this far I turned my thoughts toward when and how we should make the announcement. I knew that Ken Hulet was going to be honored at a San Diego American Chemical Society (ACS) meeting on the occasion of receiving one of the ACS awards in March of 1994 and decided that this would be an excellent time since he was one of the codiscoverers. He could announce the name during his acceptance speech. I was pleasantly surprised when I broached both ideas to him. He was not opposed to the name but he thought that Glenn would not accept it. He suggested that I ask him before we go any further in our search for a name. I said that I was willing to do so but that I thought that we should have a meeting of the seven of us and make sure that we

had unanimous agreement. I said that if I could report to Glenn that this was the case he would be much more willing to go along with it.

"The meeting took place on Tuesday, November 30, 1993, at the SuperHILAC. For the meeting I had prepared a loose-leaf notebook for each of us which was made up mostly of copies of excerpts from the logbooks concerned with the second element 106 discovery in 1974 and that in which we first observed it in 1971. I also included other pertinent documents along with the *Physical Review Letters* article that was submitted on the confirmation work at the 88-Inch Cyclotron. Everyone was pleasantly surprised; it is a great memento.

"We discussed the ground rules for picking a new name in case Glenn would not accept our proposal. We decided not to pick an alternate, because everyone agreed that Glenn should be involved if we did that. We then went around the circle of seven to make sure that we were unanimous in the big decision. Again the only dragging of feet was based on whether Glenn would accept. Both Carol and I felt certain that he would do so, because he took the long view of history. The name would last forever (whatever that means) and it was right that his should be on the roster. Jose's view was that I should not accept no for an answer.

"The next step was for me to approach Glenn. I had an extra day to think about it, since the next day Wilma and I went out birding, so I set up an appointment to see Glenn on Thursday, December 2, 1993, after lunch. I decided to let him know of our decision in an unusual way. I made up a special cover page by hand for the 'Element 106 Story' folder. It is shown in Fig. 10.7. It looked great. Explaining that I had something important to show him, I presented the folder and watched as he opened it to the first page.

"He was clearly astonished — and pleased, too. I now felt confident that he would accept our decision. We talked for a little while about the big step and I told him how it had come about. He told me that now he understood why I had been so evasive in my answers to his queries about how the naming procedure was progressing, especially when my normal attitude was very forthcoming. After the 88-Inch Cyclotron confirmation we had discussed it and I had said

Dear Glenn,

The team has decided unanimously that the only name for element 106 is yours!
Thus the roster becomes:

109	Mt	meitnerium
108	Hs	hassium
107	Ns	nielsbohrium
106	Sg	seaborgium
105	Ha	hahnium
104	Rf	rutherfordium
103	Lr	lawrencium
102	No	nobelium
101	Md	mandelbrium
100	Fm	fermium
99	Es	einsteinium
98	Cf	californium
97	Bk	berkelium
96	Cm	curium
95	Am	americium
94	Pu	plutonium

Kon Hulet will make the announcement in San Diego in March 1994 at the symposium in his honor.

Sincerely, Albert Ghiorso. 2 Dec 1993

Fig. 10.7. Cover page for the Log Book excerpts presented to Seaborg.

that I was looking into the matter of the name. Every now and then he would ask me about it and, unusual for me, I would make some noncommittal remark. I told him that I had asked the opinions of Darleane and Paul Karol and I told him of the Markowitz independent suggestion. After about a half hour he said that he would like to think about the idea overnight and talk to Helen and he would let me know tomorrow about his decision.

"The next morning I called him and asked about Helen's reaction. He was jubilant and said that she was ecstatic. He said that he was so excited that he had sat down last night and read the whole folder that I had put together. We talked a little in general terms — and then he told me that this was the greatest honor that he had ever received in his life! Coming from him, this statement made me feel great. We did the right thing."

It was then decided by the discoverers that Ken Hulet (LLNL) would announce their decision at the meeting of the ACS in San Diego at the opening symposium on Sunday, March 13, 1994 to honor Hulet's receipt of the prestigious ACS Award for Nuclear Chemistry. His dramatic announcement was met with surprise for very few people knew in advance about it. Then, as the full import of it was realized, the audience erupted with extremely enthusiastic and long-lasting applause. It was a great moment which will long be remembered.

Later, Dr. Ken Gregorich, a former Ph.D. student of Seaborg, a postdoc with Hoffman and then a Nuclear Science Division Fellow in Hoffman's group in the Nuclear Science Division at LBL, described Hulet's many contributions to actinide science. Then he outlined the results of the seaborgium confirmatory experiment.

The name 'seaborgium' for element 106 was endorsed by the ACS Committee on Nomenclature, and by the Board of Directors shortly thereafter. After some intervening initial disparate recommendations by the IUPAC Commission on Nomenclature of Inorganic Chemistry, approval finally came at the meeting of the IUPAC Council in Geneva, Switzerland, on August 30, 1997. Figure 10.8 shows Glenn pointing to his element.

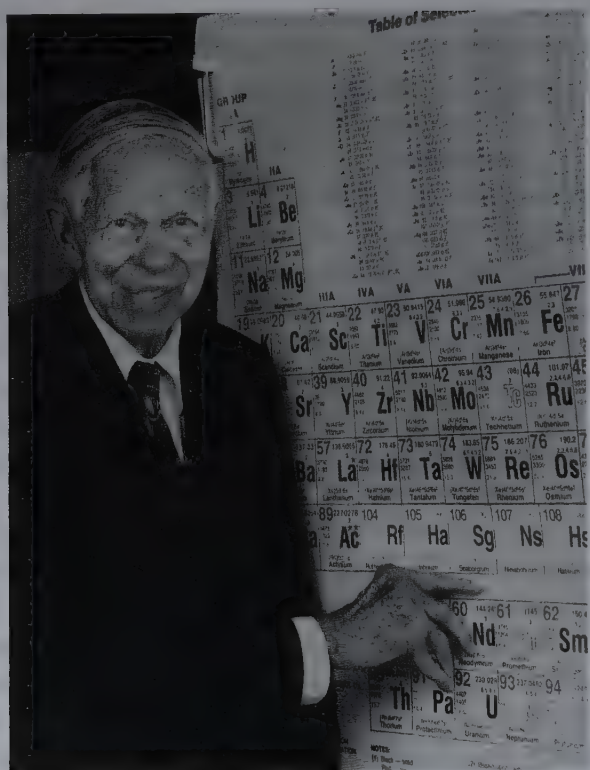


Fig. 10.8. Glenn T. Seaborg pointing to seaborgium on the periodic table of the elements.

10.4. First Studies of Chemical Properties of Seaborgium

Recently, the longer-lived isotopes ^{265}Sg and ^{266}Sg , with half-lives of about 7 and 21 s, respectively, have been synthesized and identified.^{10.6-10.9} This has made it possible for an international collaboration including researchers from the USA, Germany, Switzerland, Norway, Sweden, Japan, and China to study both the aqueous and gas phase properties of seaborgium.^{10.7-10.9} In general, seaborgium has so far been found to exhibit the properties expected of an element of group 6 of the periodic table. In preliminary experiments

of its sorption on an anion exchange resin column, seaborgium was found to run through like the heavy group 6 homolog tungsten rather than sorb like U(VI), which forms the positively charged uranyl ion. In the gas phase studies,^{10,9} it appears that Seaborgium forms SgO_2Cl_2 , similar to its lighter homologs chromium, molybdenum, and tungsten. Thus it seems that we can look forward to a variety of seaborgic and seaborgous halides and seaborgate compounds as well.^{10,10} Although seaborgium appears to belong to group 6 of the periodic table, it is not yet clear whether its properties can be smoothly extrapolated from its lighter homologs and more detailed studies will be required to determine whether it behaves more like tungsten or molybdenum.

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Chapter 11

Bohrium (107), Hassium (108), and Meitnerium (109)

11.1. Introduction: The UNILAC

GSI owes its existence to two fortunate circumstances: the development of a new type of accelerator and a campaign by physics professors in Hessen to set up a joint center for university research. The result was a research center of international standing. Up to this point Germany had been following the customary practice used throughout the world which can be characterized as "to each institute its own accelerator." As a result, since the mid-50s some 20 small to medium size commercial accelerators — tandems, cyclotrons, Van de Graaffs, and electron linacs — had been installed in the Federal Republic for use in low energy spectroscopy. By and large these were low energy accelerators and thus were limited mostly to the lower end of the periodic table for nuclear physics experiments. By 1966, scientists at Marburg and Frankfurt were in the planning stages for the next logical step to increase the energies of these small machines by using what were sometimes called "afterburners." There was nothing available commercially, so they set out to design and construct their own devices. They were competing groups, each with its own idea as to how best to accomplish its goal. But they also knew that in unity there was strength, so the university lecturers in the state of Hessen set up a nuclear physics study group, Kernphysikalische Arbeitsgemeinschaft Hessen (KAH), with the aim of coming together to establish a larger central accelerator laboratory for their physics institutes instead of continuing the practice of each

having its own, smaller unit. Inspired by the old idea of uniting research and teaching, the intention was that the laboratory should aid research at an institute of excellence and at the same time promote the education of students.

Now an amazing thing happened. After a little "brainstorming" Wilhelm Walcher, Erwin Schopper, and Peter Brix decided to put aside their own local interests and join forces to really strive for a joint center. This was a courageous move but one which also showed great perspicacity. They knew that a cooperative endeavor would benefit everyone, because in this way they would all share a better and more powerful machine. They knew that modern science now demanded accelerators that were much more complicated and thus expensive so that some sort of joint venture would be needed. The big question was, What kind of accelerator should it be and where should it be located?

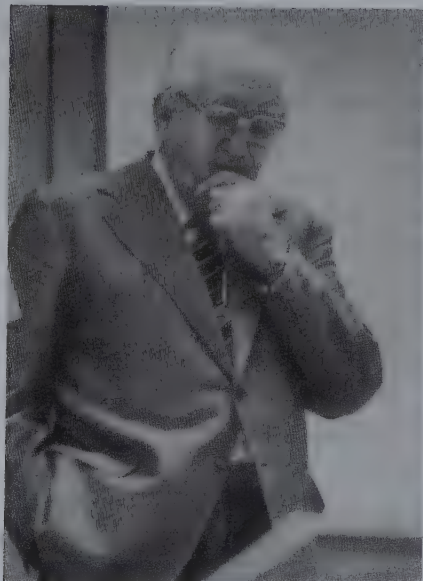


Fig. 11.1. Prof. Dr. Christoph Schmelzer, founding father of the UNILAC at the ceremony to name elements 107, 108, 109. September 7, 1992.

In the late 50s, Christoph Schmelzer (Fig. 11.1), who at the time was working in Geneva on the development of the CERN proton synchrotron, had considered possible ideas for a linear accelerator for ions all the way up to uranium. Schmelzer was much ahead of his time, because in those days even carbon and oxygen ions were regarded as "heavy." Following his appointment to the University of Heidelberg, he systematically pursued the development of this type of accelerator. His UNILAC (UNiversal Linear ACcelerator) was to be a high frequency, variable energy accelerator, capable of accelerating ions of all elements to energies above the Coulomb barrier. Given that the "accelerator world" of the time consisted basically of "off the shelf" tandems and cyclotrons, this was a bold and farsighted venture.

It is interesting to note that these new ideas in Germany appeared at about the same time that the ill-fated Berkeley Omnitron project was being pursued. The Berkeley Omnitron was a more ambitious and more versatile accelerator but, unfortunately, it never survived (see Chapter 10, pp. 304–307). If it had survived it would have been a worthy competitor to the UNILAC and would have come on-line several years before that machine and a couple of decades before the storage ring ion source (SIS), the high energy addition at GSI.

Christoph Schmelzer's machine quickly won out over its competitors when he joined the KAH project in 1967, and as the scale of the enterprise grew, so too did insight into what it involved. The KAH, which originally had been limited to the small area of Hessen, had now spilled over into Heidelberg and Mainz and the project had rapidly outgrown the role of a single establishment funded only by that State. Now the aim was to acquire the federal government as an additional sponsor of this laboratory alongside the State. In other words the project was to become a major research center — similar to Deutsche Elektronen Synchrotron (German Electron Synchrotron, or DESY), which had been founded in Hamburg in 1959. In addition, the neighboring universities were developing experiments and equipment highly relevant for heavy ion research. These included mass separators at Giessen and Marburg, detectors at Frankfurt,

Heidelberg, and Marburg, as well as nuclear chemistry techniques at Darmstadt, Mainz, and Marburg. All these developments helped to give a more varied structure to the scientific program, and thus heavy ion physics became generally accepted as the scientific goal of the new center from the earliest stages. The GSI was officially constituted as the "Gesellschaft für Schwerionenforschung mbH" on December 17, 1969. It had the legal form of a private limited company and the status of a "Großforschungseinrichtung" — a major research center — of the federal government and the state of Hessen. The research center was headed by a five-member management committee, the Scientific Directorate — Prof. Rudolph Bock, Prof. Gisbert zu Putlitz, Prof. Peter Brix, Prof. Christoph Schmelzer, and Hans Otto Schuff, as the administrative director. In keeping with the founding fathers' idea of "proximity" to university interests, the university groups were involved in the preparation of experiments from the planning stage onwards and, accordingly, a new type of funding model — later known as the GSI model — was established. It incorporated a budget allowance for the university groups so that they could develop methods and equipment, and finance staff. This budget item initially amounted to over 70% of GSI internal research funds and still accounts for around 30% of these funds today. Thanks to such generous funding, the university groups were able to quickly get started on the big job of putting the broad research program into effect.

The construction of the UNILAC began shortly after GSI was founded at Wixhausen, near Darmstadt, West Germany (see Fig. 11.2). At the time, important arguments for choosing this type of accelerator included the fact that the particle energy could be varied readily and the fact that injection could be easily achieved from a range of ion sources. These characteristics also proved to be very valuable when the installation was later expanded. Before construction could begin, however, much basic research into the design of the machine had to be done, because there were many new ideas that had been incorporated in the plan; thanks to the prior work at the Institute of Applied Physics in Heidelberg this design work went swiftly and in



Fig. 11.2. Gesellschaft für Schwerionenforschung mbH (GSI Laboratory) group, Darmstadt, Germany. Peter Armbruster, Christoph Schmelzer, Rudolph Bock, Hans Otto Schuff, Helmut Ahrens, Glenn T. Seaborg, Gunter Herrmann, and Hans Floeter, September 1972.

November 1975, after just four years of construction, the Center's UNILAC produced its first heavy ion beam. Part of the experimental setup had already been completed, so the accelerator was immediately put to use, but there were still many challenges that had to be met, one of the most important being a shortage of trained people in the field of accelerator electronics and operation. These people learned "on the job" during the phase of what is sometimes called the "conditioning" of the accelerator. Mastering the challenges of improving the quality of the beam and developing sources capable of producing the broad spectrum of ions required was to occupy the accelerator team completely over the next few years.

11.2. Cold Fusion

A major development which led directly to the discovery of elements heavier than Seaborgium was the concept of what has become known as "cold fusion." In 1974 Yuri Oganessian pointed out that one can take advantage of the Q values for heavy ion reactions to

reduce the excitation energy to a minimum. In practice, this meant the use of lead and bismuth targets with medium-heavy ions rather than the use of transuranium targets with lighter ions. By choosing the proper partners the number of particles that had to be evaporated to remove this excitation energy was minimized. The effect was to reduce competition from fission and, consequently, increase the cross section for the desired heavy element. The principle was tested successfully by Oganessian and his group^{11.1} with reactions that produced fermium and transfermium isotopes, and it should be noted that the discovery of this principle was one of the keys to much of the heavy element work beyond seaborgium.

It is strange that the Dubna people did not really exploit their discovery of cold fusion. Instead, they stayed rooted for two decades in the use of SF as their only viable detector of the fusion products. SF is an important mode of decay but is quite limited in the information that it provides, whereas alpha decay is quite definitive even in the case of a single atom, and they must have appreciated this. In the early days their actions were probably dictated by the fact that Flerov was the codiscoverer of spontaneous fission and they were very familiar with that mode of decay. The detection of fission fragments could be done very efficiently by passive means and did not require computer technology. On the other hand, in the early 60s, Zager and Mikheev and Donetsk used modern silicon detectors in various separate experiments to make good alpha-particle measurements of the isotopes of nobelium and these were on a par with those of the Berkeley group. Why were these same techniques not applied to the heavier elements just as they were in Berkeley?

11.3. SHIP

Just as important as the recognition of cold fusion was the fact that it took very specialized devices to detect and identify these new elements. The first of the new sophisticated instruments to be constructed at GSI was the SHIP (Separator for Heavy Ion Reaction Products) (See Fig. 11.3.) (The acronym was knowingly picked,

SEPARATOR FOR HEAVY ION REACTION PRODUCTS (SHIP)

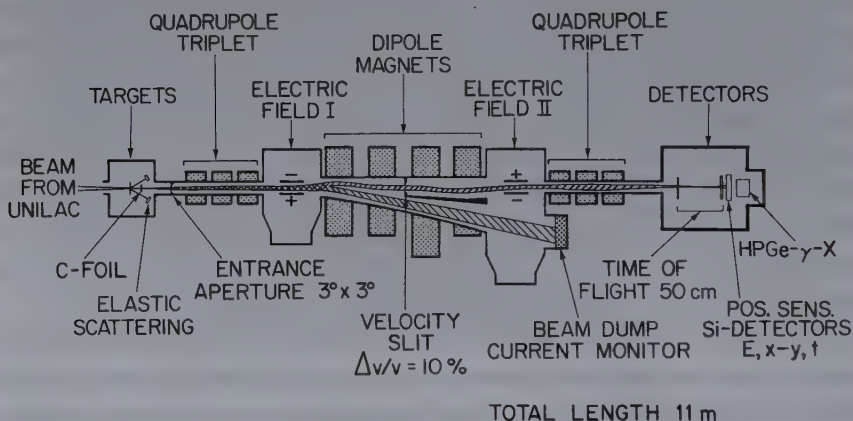


Fig. 11.3. Schematic of SHIP.

because Armbruster knew that they were going to embark on a discovery mission across an unknown Sea which, hopefully, might take them to the fabled Magic Island of the SHE!) This instrument, based on work at Giessen, was built over a period of a number of years under the direction of Peter Armbruster in the 70s. It is a recoil velocity separator designed to separate completely the fusion products emitted in heavy ion reactions from the intense beams that produce them. The process of separating the recoil products from the bombarding beam takes advantage of the fact that the fusion products travel much more slowly than the beam particles. SHIP can be described as a two-stage velocity filter, the principle of which dates back to the German physicist Wilhelm Wien. The filter employs a combination of electrical and magnetic deflecting fields which have been arranged so that the electrical and magnetic deflections cancel each other out for a specified particle velocity; thus, the low velocity recoils are steered on the axis of the accelerator to the detector system while the beam is deflected to one side.

The velocity selection window provided by the separator is fairly narrow, approximately $\pm 5\%$ in width, so it is already a fairly good filter, but this is supplemented for each particle that passes through it by a time-of-flight (TOF) measurement. With this value and the recoil energy an approximate mass measurement for each recoil can be calculated. The TOF is measured, just in front of the focal plane, by using microchannel plates to detect the secondary electrons emitted from thin carbon foils 30 cm apart. At the focal plane the recoils are implanted into an array of position-sensitive silicon surface barrier detectors to determine each particle's point of impact and energy. Six other identical strip detectors are arranged in the form of a box with one side open so as to capture the α -particles that escape from the focal plane strip in the backward direction.

The time, energy, and position of the subsequent events that take place after an implantation are recorded to obtain precise identification of a particular implanted nucleus. This process is referred to as the genetic correlation method, because the decay products of an implanted parent nucleus have the same spatial coordinates in the detectors and can thus be correlated with one another. The α -particle decay of one of the new elements can thus be unambiguously correlated with the already known properties of the following generations of daughter isotopes. Decay chains have been observed down to the fifth generation. Gamma and X-ray Ge detectors are placed directly behind the focal plane detectors to provide further information about excited states or *ec* decay.

The total system was quite complicated and demanded the dedicated ingenuity and hard work of many scientists and technicians to construct it. But that was just the beginning, because as soon as it was shown that SHIP was a superior facility that had the potential for extending the periodic table to new heights, a continuing program was initiated to improve the separator for this goal. This demanded new insights and they were forthcoming from the young skilled team that Armbruster had assembled. Always the driving force, Gottfried Münzenberg tirelessly directed the team in the construction, debugging, and operation of SHIP.

One of the big problems encountered from the start was that it was possible for some unwanted scattered particles from the UNILAC to pass entirely through the separator because SHIP was on the zero degree line of the accelerator. Some years later this was changed with a small bending magnet to deflect the recoils 7° away from this axis, but for the first years of operation the experimenters had to surmount this problem in other ways. The simplest stratagem was to only analyze the α -radioactivity from the recoil atoms that were implanted in the detecting crystals during the beam pause, the interval between the beam pulses. Even during the beam pulse, however, most of the time they could differentiate a heavy recoil from a SHIP pulse by using the TOF to make a rough mass measurement.

11.4. Bohrium (Element 107)

In 1976, Oganessian and coworkers reported^{11.2, 11.3} the production of an SF activity with a half-life of ≈ 2 ms from the $^{209}\text{Bi}(^{54}\text{Cr}, 2n)$ reaction, which they attributed to $^{261}107$. They used a rapidly spinning target wheel exposed to the ^{54}Cr beam to carry the short-lived atoms next to passive SF detectors, but this type of simple experiment conveyed very little information and the assumption had to be made that the atomic number of the reaction product they detected had an atomic number that was simply the sum of the atomic numbers of the target and the projectile. As will be seen by the later GSI results, this assumption is often unwarranted. It was a good exploratory experiment but lacked any proof of the atomic number of the species that was made.

In 1981, Gottfried Münzenberg *et al.*,^{11.4} using SHIP at GSI with the same target and projectiles, identified the nuclide $^{262}107$, produced by the emission of only one neutron. The mass number of a velocity-separated product nucleus was roughly determined by measuring its TOF and the energy deposited in a detecting crystal; its atomic and mass numbers were determined exactly by observing the genetic relationship between $^{262}107$ and its known α -decay descendants.

Five decays of $^{262}107$ were observed with $E_{\alpha}=10.4$ MeV and $T_{1/2} \approx 5$ ms. One sequence of correlated decays ended in the known nucleus ^{254}Lr , one ended in ^{246}Cf , two ended in ^{250}Fm decay, and one ended in ^{250}Md . The cross section for producing these nuclei was $\approx 2 \times 10^{-34}$ cm² or 200 pb. (Note that this is only 2×10^{-7} of the production cross section observed in the first one-atom-at-a-time discovery experiments with mendelevium at Berkeley!) It is a tribute to the quality of the GSI experiment that its results found rapid, universal acceptance despite the low production rate involved. By 1988 a total of 38 atoms had been observed. Subsequent experiments identified three element 107 species, $^{261}107$ ($T_{1/2}=11.8$ ms; $E_{\alpha} \approx 10.2$ MeV), $^{262}107$ ($T_{1/2}=102$ ms; $E_{\alpha} \approx 9$ MeV), and $^{262}107^m$ ($T_{1/2}=8.0$ ms; $E_{\alpha} \approx 10.3$ MeV). Contrary to the initial observations of the Dubna group, no SF activities with $T_{1/2}=1\text{--}2$ ms were observed. The acknowledged discoverers of element 107 gave it the name “nielsbohrium” (symbol Ns) in honor of the great Danish physicist Niels Bohr (Fig. 11.4), the architect of atomic structure, but this was later changed to “bohrium” (symbol Bh) by the IUPAC in 1997 (see Chapter 13). The name “nielsbohrium” had been suggested

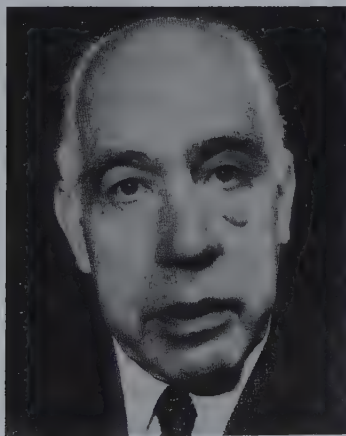


Fig. 11.4. Niels Bohr (1885–1967).

earlier for element 105 by the Dubna group, but GSI asked them for permission to use it for element 107 in order to recognize Oganessian's contribution in suggesting the use of cold fusion reactions to prepare elements beyond 106.

11.5. Hassium (Element 108)

In 1984 a report of the successful synthesis of element 108 appeared. The Darmstadt group used SHIP^{11.5} to identify three atoms of element 108. They used the nuclear reaction $^{208}\text{Pb} (^{58}\text{Fe}, n) ^{265}\text{108}$ at a ^{58}Fe energy of 5.02 MeV/nucleon, resulting in an excitation energy of only 18 ± 2 MeV for the compound system. The cross section for production of these nuclei was 1/10 that observed for the production of element 107. Three time-correlated α -decay chains that clearly led to the known daughter nuclei, $^{261}\text{106}$ and $^{257}\text{104}$, were observed. The new nuclide, $^{265}\text{108}$, had a $T_{1/2} \approx 1.8$ ms and decayed by the emission of 10.36 MeV α -particles.

In a second experiment,^{11.6} one atom of the even-even nuclide $^{264}\text{108}$ ($T_{1/2} \approx 80$ ms) was produced in the $^{207}\text{Pb} (^{58}\text{Fe}, n)$ reaction. The observation of α -decay by an even-mass number isotope of element 108 was an immensely important sign that SF lifetimes were unexpectedly long for these nuclei, and boded well for the synthesis and identification of still heavier nuclei. (It is interesting to note that the element 109 experiment was conducted before that for element 108 because it was thought at that time that it was likely that $^{265}\text{108}$ would decay by SF rather than by α -emission, and if that were the case, there would be no way to determine the atomic number of the spontaneously fissioning element 108 based on detection of its fission products.)

At about the same time, Oganessian *et al.*^{11.7} reported the observation of the possible decay of $^{263-265}\text{108}$ produced in the reactions $^{209}\text{Bi} (^{55}\text{Mn}, n) ^{263}\text{108}$ and $^{207, 208}\text{Pb} (^{58}\text{Fe}, n) ^{264, 265}\text{108}$. The production cross sections reported by this Dubna group were only 1/10–1/4 those observed by the Darmstadt group for element 108.

None of the α -particle decays of these nuclei were observed directly. In the case of $^{264}_{108}$, an 8-ms and a 6-ms SF activity were observed and attributed to $^{256}_{104}$, the known 7-ms SF activity granddaughter of $^{264}_{108}$. Similarly, $^{263}_{108}$ was identified by the observation of a 1.1-s SF activity attributed to its granddaughter, $^{255}_{104}$, a known 2-s SF activity. The nuclide $^{265}_{108}$ was claimed to have been detected because of the observation of α -emitting $^{253}_{83}\text{Es}$, a possible great-great-great-granddaughter of $^{265}_{108}$. Interesting as the observations of the Dubna group are, they are hardly sufficient by themselves to be a claim for the discovery of element 108.

The acknowledged Darmstadt discoverers of element 108 have given it the name "hassium" (symbol Hs), in recognition of the German state of Hesse (Latin: Hassia), where the GSI Laboratory is located. This name was officially adopted by the IUPAC in 1997 (Chapter 13).

11.6. Meitnerium (Element 109)

In 1982, Münzenberg *et al.* reported^{11.8} the observation, during two weeks of bombardment, of one unusual time-correlated decay sequence for the reaction product, $^{266}_{109}$, that had been velocity-separated by SHIP from the $^{209}_{83}\text{Bi} + ^{58}_{26}\text{Fe}$ reaction. Such a yield corresponds to a formation cross section of ≈ 10 pb. After implantation of the complete fusion reaction product in a detector, an 11.1-MeV α -particle was detected, followed 22 ms later by a 1.14-MeV α -particle, followed 13 s later by SF. The 1.14-MeV energy signal is assumed to result from an α -decay in which only part of the energy was deposited in the detector.

In a second experiment in 1988 two new time-correlated decay sequences compatible with the first event were found.^{11.9} The combined results of the two experiments for $^{266}_{109}$ gave a value of the half-life of ≈ 3 ms and a production cross section of ≈ 10 pb. A third experiment in 1997 mapped out a complete excitation function with approximately 20 events.



Fig. 11.5. Lise Meitner (1878–1968).

The acknowledged GSI discovery group gave element 109 the name “meitnerium” (symbol Mt) in honor of Lise Meitner (Fig. 11.5), which the IUPAC officially approved in 1997 (Chapter 13).

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Chapter 12

Elements 110, 111, and 112

12.1. Element 110

12.1.1. *Berkeley's SASSY2*

It is notable that LBL had been the pacesetter in producing new elements from element 93 up through element 106, seaborgium, and yet this success did not bring about the continuing financial support that would allow the construction of the more elaborate instruments needed to go ever higher in atomic number. Mike Nitschke had proposed an elaborate vacuum recoil spectrometer for use at the SuperHILAC in the 1980s that was very much like SHIP. It was intended for all of the users of that accelerator as well as for heavy element research but, unfortunately, it did not win the wholehearted backing of the "powers that be," both national and local, because of competition from the BevaLAC; it only got as far as the construction of the large concrete pad which joins two buildings and today is used as a parking lot. It is ironical that the heavy element program which was completely responsible for the creation of the BevaLAC and its subsequent expensive research program should be ultimately sabotaged by the voracious needs of that program!

Fortunately, the attempt to kill the new-elements research effort was not completely successful and in August–September 1991, the element 110 experiment, the last to be performed at the SuperHILAC, was undertaken by a large group. In spite of the many difficulties that were encountered, the long experiment was successful, in that a single candidate event with many of the expected characteristics of $^{267}_{110}$ was observed. Although we regard the event as significant,

whether it was assigned correctly can only be determined by some future experiment.

After the discovery of seaborgium in 1974, it was clear that detecting heavier elements would demand the use of fairly sophisticated instruments because of the short half-lives and very small production cross sections that were expected; what was not clear was how best to separate them from the intense heavy ion beams that would be needed for their production. Ghiorso, on the basis of his experience with the Omnitron design work, realized that it should be possible to take advantage of "charge focussing" in a gas-filled magnetic field. This term refers to the fact that, although a recoil ion traveling through a low pressure gas experiences multiple charge states, on average it acts as if it has only a single mean charge state because of the many up and down iterations of charge states in each millimeter of its path. This characteristic, which varies with Z and velocity, causes the slow heavy recoiling ions to have a larger magnetic radius of curvature than the fast ion beams that produce them and thus offers an inexpensive option for an instrument to isolate them. The earliest exploration at Berkeley using the Jupiter steering magnet at the end of the Post-Stripper tank of the old HILAC showed that the gas-filled principle did have good potentialities; the problem was how best to translate this idea into a practical device for experimental use. Initially, conversations with Klaus Halbach, who had been important in the Omnitron project and later invented the extremely important wiggler concept for the synchrotron light sources, were invaluable in setting the group on the right path to designing a suitable magnetic system.

Subsequently, computer analysis by Saburo Yashita, one of Seaborg's graduate students, with the assistance of Juris Kalnins of the Bevatron staff, resulted in a design which was known by the acronym SASSY (Small Angle Separator System) (Fig. 12.1). Saburo was a very creative Japanese student who had worked with Bill Libby at UCLA as an undergraduate chemist for two or three years and now was working under Seaborg to earn his Ph.D. in the heavy element program. He was assigned to work with Ghiorso, who is not

SMALL ANGLE SEPARATING SYSTEM (SASSY)

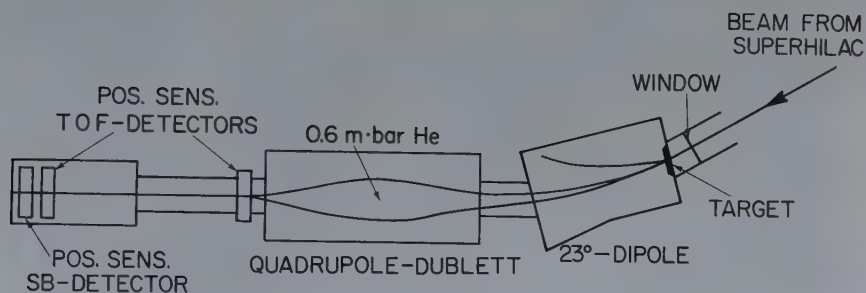


Fig. 12.1. Small Angle Separating System (SASSY).

a chemist, so perforce Saburo became a physicist and an engineer while he was being "Americanized." After completing his computer design, Saburo and Kalnins went "scrounging" around the Hill for suitable magnets that could be the basis for SASSY. They ended up with a large C-magnet and an 8-inch quadrupole doublet from the Bevatron stockpile that would bend the heavy fusion recoils emitted from the target through 30° and bring them to a focus. After many months of assembling these magnets into a workable separator, it was tried out with heavy ion beams from the SuperHILAC with targets in the lead-bismuth region to make new astatine and polonium α -emitters.

As expected, there was a very large separation of the recoils from the beam, though not from target-like recoils, in a distance of 4 m at a pressure of 1 torr of helium before the recoils were implanted into a smallish solid state detector array. For experiments where the cross sections were large (10^{-32} cm²) it was a good instrument and Saburo and Matti Leino, a young visiting scientist from Finland, were able to discover several new isotopes and use this research as thesis subjects. The heavy elements were another matter, because their cross sections would be a thousand times smaller, so it was decided to add a TOF capability to enable the slow fusion recoils to be

distinguished from the fast target-like products. This turned out to be very difficult to do because of the ambient 1-torr pressure of helium that was needed for the separator, and although we were successful the final results were well short of expectations.

In 1982 a collaboration was set up between GSI and LBL to make a search for the Super Heavy Elements (SHE). It was proposed to undertake joint experiments both at Berkeley and Darmstadt with ^{48}Ca bombardments of ^{244}Pu using both physical and chemical techniques to identify them. To further the cooperation between the two laboratories, there was an exchange of some of the personnel in 1982 and the following year to prepare and conduct the experiments. Peter Armbruster and Peter Lemmertz came to Berkeley for a period of several months and worked closely with our people to help solve the many problems that were being encountered with SASSY. The following year Ghiorso went to GSI for a few weeks to assist in the SHIP experiments with ^{48}Ca on ^{244}Pu . At Berkeley Peter became fascinated with our separator, since he had used the same principle for separating fission products with his device called Josef, which he had constructed at the Research Center at Jülich. The problems encountered now were new and quite different, however, and their solutions induced Peter to assemble a scholarly paper on SASSY^{12.1} which was published several years later. Other members of the Armbruster group, J.-P. Dufour, Gottfried Münzenberg, and Sigurd Hofmann, also came to Berkeley for short periods of time to assist in preparing our instrument for bombardments. After a long development period SASSY was used successfully in a ^{48}Ca bombardment of ^{208}Pb to detect the 55-s α -emitter, ^{254}No , made in a 2-n out reaction. Somewhat later it was used in unsuccessful searches for SHEs in bombardments of ^{244}Pu and ^{248}Cm . Figure 12.2 shows some of the members of the LBL/GSI collaboration.

These first results were welcomed and they did show that the principle was sound. However, they also showed that if the fabled Super Heavy Element Island was to be reached, the separator would need a larger acceptance angle to accommodate ^{48}Ca fusion reactions and better magnetic resolution to free the focal plane from transfer



Fig. 12.2. Mike Nitschke, Matti Leino, Al Ghiorso, Ken Moody, Peter Armbruster, Peter Lemmertz, Sigurd Hofmann, Saburo Yashita, Gottfried Münzenberg, and J.-P. Dufour at SuperHILAC, LBL, 1982.

products. Accordingly, a few years later SASSY2, a new double-focussing gas-filled magnetic separator with a larger bend angle and a shorter path length was designed by Kalnins to take care of these shortcomings; it was of the dipole–quadrupole–dipole (DQD) type, where each dipole had a strong magnetic gradient. Instead of trying to use our difficult TOF system, it was decided to compromise and use a CO_2 -filled energy loss proportional detector to make a rough measurement of Z in the space between the end of the magnetic field and the focal plane detector.

After GSI had shown the way in the new element field up to element 109, the question naturally arose as to which nuclear reaction we should use to try and make element 110. From a theoretical standpoint it was generally agreed that the optimum reaction to make element 110 was $^{208}\text{Pb}(^{64}\text{Ni},n)^{271}110$, but this nickel isotope was rare and thus very expensive and not available to the Berkeley

experimenters. If we had been able to use this isotope the cross section would have been 10–20 times higher and the lead targets would have stood up to our maximum beam better than did the bismuth targets. The only option was to use the commonly available isotopes and bombard ^{209}Bi with ^{59}Co ions at an energy near the barrier and hope to observe the production of $^{267}110$ after the emission of one neutron. This nucleus was expected to be a very short-lived α -particle emitter (predicted energy 11–12 MeV) that would decay to known nuclei of lower atomic numbers. The production cross section was expected to be in the neighborhood of a picobarn and we knew that if everything went well this goal was within reach.

There was one major flaw in this plan — how were we going to construct this new separator? An adequate budget was not available from the LBL Nuclear Science Division (NSD) to build the equipment that we would need: the separator, the rotating target wheel system, the 50-crystal focal plane detector, and the complicated electronics array that would store and decode the information from the detector. Unfortunately, the management no longer gave a high priority to the heavy element program in spite of the fact that it was this program that had been largely responsible for bringing it to its present prominence. We knew that if we tried to launch a project of this magnitude it would be denied, so we decided to do what used to be normal in the first decade or two after Lawrence's invention of the cyclotron — i.e., do it yourself!

The magnetic system was basically a big milling job and though some aspects of the device were very large and challenging, i.e., milling the large pole pieces into the complicated shapes needed, it was largely constructed and tested by Ghiorso, himself, with major help from his son, Bill (working on his own time). Bill's regular job at the Laboratory was as an engineering technician for the Heavy Ion Fusion Project. Together, they slowly constructed SASSY2 (Fig. 12.3), solving problems as they appeared. One of the important ones was how to protect the SuperHILAC against an explosive breakage of the thin window that isolated the 1-torr helium environment of SASSY2 from the accelerator. Bill solved that one by designing a fast

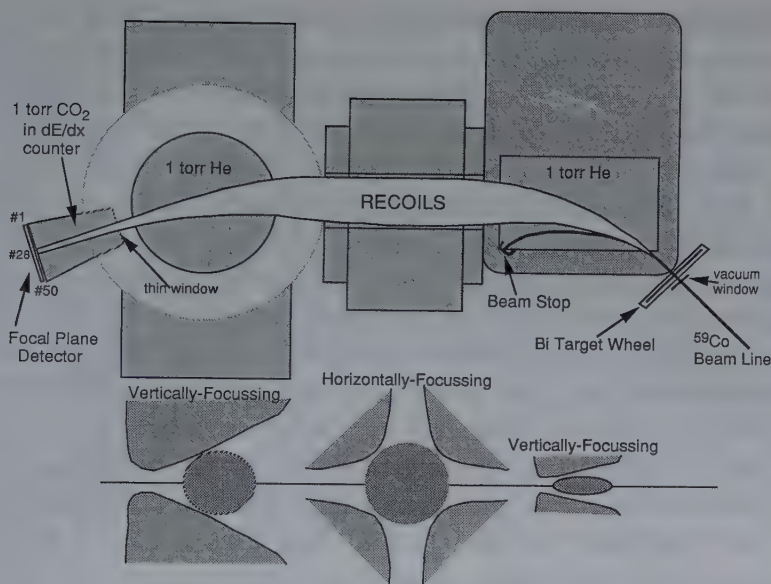


Fig. 12.3. SASSY2.

"slammer" valve powered by the spring of a large rattrap! This was vital because the accelerator was pumped cryogenically with helium at 20°K and thus had no capability of pumping helium at all. Bill also designed and built the driver system for the target wheel and other necessary electronic gadgets needed for the experiment. After completion it turned out that it was very difficult to align the separator, because, although it had a DQD configuration, the dipoles had strong quadrupole components so that it was really three quadrupoles in series. Since the usual elaborate magnetic measurements survey that is normally used for alignment purposes was not affordable, it was decided to use instead an α -source; this turned out to be a cheap and effective tool. When he could spare the time from his usual accelerator technician duties devoted to keeping the SuperHILAC going, Don Syversrud was extremely valuable in assisting with the alignment and giving critical advice and assistance in the construction of SASSY2.

The detector system at the focal plane consisted of a large area 50-crystal position-sensitive silicon detector array built by Jack Walton at LBL, since at the time of the experiment there were no large area detectors available commercially. The detector system needed many high quality amplifiers for recoil, α -particle, and SF detection as well as a host of other electronic gadgetry needed for a working system. At that time it would have been prohibitively expensive for our meager budget to buy commercial units, so Alfred Wydler designed his own complicated system. It worked rather well, its only drawback being that if anything went wrong only Wydler could fix it! The data acquisition programs were designed and debugged by Richard Leres working with Wydler. The two worked closely as a team to solve the complicated problems that always seem to arise when electronics has to mesh with computers. The third member of the team that handled the data stream was Diana Lee, a longtime scientist, engineer, technician, and collaborator of Darleane Hoffman. Diana was invaluable, during the startup of the detector equipment when we had to learn the idiosyncrasies of the system and especially when it came time to decipher the information that we had obtained from the big run.

Normally, there would have been a long commissioning period with beams from the SuperHILAC to determine the characteristics of SASSY2 *accompanied by suitable periods to correct any defects*, but, as it happened, what little beam time we were able to obtain was completely inadequate. This was because the BevaLAC's need for the SuperHILAC as an injector was always given an absolute priority, and in practice this meant that a tuning that would help a low energy beam for a SuperHILAC user was not allowed if it interfered with a beam destined for the BevaLAC. When the BevaLAC needed uranium ions no other ion could be used in the time share mode, because uranium acceleration was quite difficult for the accelerator. When everything was just right the time share option worked well, and if one of the other SuperHILAC users did not need the beam it could be directed into our separator. Most of the time the opportunities to use the beam were limited in their usefulness.

(It was at times like these that Ghiorso would curse ever having invented the BevaLAC!)

Matters were brought to a head on July 8, 1991, by a memo from the Scientific Director for the low energy program of NSD, terminating the SASSY2 program and the element 110 experiment as of September 1, 1991. He stated that the most recent budget crunch had forced program cuts and he and his Scientific Advisory Committee had given the lowest priority to the search for element 110.

Seaborg and Ghiorso then met with the NSD management to try to persuade them that the element 110 experiment should be allowed to run. After an hour's "heated" discussion it was agreed that the experiment would go ahead until the end of the fiscal year, September 30, 1991, but if unsuccessful, that would be the end of SASSY2. The result was that now there was the chance that if everything worked properly the bureaucracy would be shown to be wrong in its judgment!

We intensified our preparations for the Big Run and gathered together additional people into a temporary loose organization called the "Coalition," to participate in the search. The "outside" group included Torbjorn Sikkeland (retired) from Norway, Saburo Yashita from Monaco, Andy Türler from Switzerland, Matti Nurmi from the US and Finland, and Walter Loveland from Oregon State University. Hoffman's graduate students, especially valuable for shift duty, and various other people from Berkeley made up the bulk of the list. It was a great reunion of people who had worked in the heavy element field. There would have been others except for conflicting commitments.

The time allotted for our last run was from August 23 to September 23, 1991. This was a woefully inadequate amount of time for the experiment considering that SASSY2 had not really been tested properly. Ghiorso decided that, nonetheless, we had to devote some part of the beam time to making stand-in radium and actinium α -activities so that the system could be calibrated and its operation checked. We chose to use ^{51}V bombardments of terbium and dysprosium for this purpose. The tests with ^{51}V began on August 25

using the Adam injector and almost immediately we ran into problems with SASSY2, but to our surprise we found that the accelerator also had problems that had not been noticed before.

There were several difficulties with the SuperHILAC or its auxiliary equipment that came to light in the course of the experiment:

(1) Early on, it was discovered that the normal phase probe measurements of the beam energy that were routinely made by the accelerator staff were sometimes in serious error. After discovering this near the beginning of the big run we went to some trouble to independently monitor the energy with our own crystals, since the correct energy was crucial to what we were trying to do. This led to our setting up an experiment using one of Walter Loveland's special crystals that he had previously calibrated for pulse-height defect corrections at the 88-Inch Cyclotron. He repeated these measurements at the Cyclotron after the SuperHILAC run.

(2) Most surprising was the discovery that the Faraday cups that measured the beam intensity of the final accelerated beam were all reading too high by a factor of 2, at least when measuring an ion as heavy as ^{59}Co ! This was such a surprise to everyone that Matti Nurmia set up a calorimeter experiment and independently confirmed that finding from the crystal measurements. Matti is an expert in many branches of science and has often come up with creative solutions to problems that have arisen in his experimental work at Berkeley. The reason that this was important was that we had to know whether the ^{59}Co beam was being delivered efficiently to SASSY2. (The Faraday cup problem turned out to be caused by insufficient field strength of the permanent magnets used to suppress secondary electrons.)

(3) After a series of window failures we became suspicious that there might be something wrong with the new electrostatic beam wobbler, so Mike Nitschke, who had just returned from a European trip, investigated and found out that one of the power supplies was synchronized with 60 Hz and that was the reason that the wobbler was not doing its job as well as we thought it should. He and Phil

Wilmarth solved the difficulty by building two new supplies operating at 400–500 Hz as a substitute and we were able to see a substantial improvement.

(4) Some circuitry had been set up to turn off the beam in microseconds as soon as a recoil entered the focal plane detector with about the right energy and a coincident dE/dX signal from the proportional counter next to the focal plane. This would have eliminated entirely the small background that occurred while the beam was turned on and thus allowed us to look for daughter, granddaughter, etc. events with the lowest possible interference. Unfortunately, the high voltage circuitry to gate the beam off failed almost immediately and we never had a chance to use it in the actual experiment.

Although at times the accelerator difficulties were crippling, some of the problems encountered were with SASSY2 itself. Paramount among these was the single window which isolated the 1 torr of helium of the separator from the high vacuum of the beam line to the accelerator. This window was made of carbon or aluminum or combinations of the two and was very thin, typically of the order of $100 \mu\text{g}/\text{cm}^2$. It had not been tested under high intensity heavy ion beam conditions, but we had calculated that it should withstand currents of a particle microampere of heavy ions if the beam was uniformly distributed. It did stand up to intense beams but it was relatively fragile so that if the beam focus were to suddenly narrow down the foil could be destroyed. It was also likely that a single tiny dust particle in the cooling helium gas that jetted against the window could act as a missile and cause breakage occasionally. When something did go wrong and a foil broke, the targets on the rapidly rotating wheel which was adjacent to the window sometimes would also break. There were numerous opportunities for failure and there was not enough operational experience to determine and rectify all of the causes. What is known is that the window problem was too often a disastrous factor for the element 110 run.

Also unfortunate was an unforeseen problem that was never solved and that severely handicapped the experiment. Al Wydler

and Rich Leres had invented and developed a workable system for running the detector complex but it had an inherent dead-time of 280 μ s. Thus, if a recoil was implanted in a crystal detector which then decayed by emitting an α -particle these two events would be recorded by the first and second ADCs. If a third event, the daughter, took place within 280 μ s it would not be recorded at all. We did not have a multiple ADC system, so to cover this interval we had purchased a commercial device called a Transient Recorder that was intended to record trains of pulses after being triggered to handle that situation. Unfortunately, we found that we could not make the device perform in the way that we wanted it to. This was tragic and it seemed that we had no way around the problem. It had worked properly in preliminary tests without beam, but when it was hooked up to the trigger signals from the on-line electronics circuitry we could not make it respond only to the signals that we were interested in. Al and Rich used all of their wizardry to no avail and this very important part of the experiment had to be abandoned because there were other problems demanding their attention. This is an example of the kind of difficulty that could have been solved if our beam time had been spread out rather than assigned to one limited period. All of these problems took time and ingenuity to solve and were quite draining of human energy, but we managed to keep going.

The biggest blow came when we discovered that the beautiful bismuth targets which had been vaporized onto 250- μ g/cm² aluminum by Alan Lyon at LBNL, did not adhere well and began to peel off after they had been bombarded for a few days at high beam levels, probably because of flexing of the targets as the nine-target wheel rotated them through the intense beam. There was nothing that Alan could do in the short term about this, so the allowable beam level had to be reduced.

The team plowed on in spite of the many difficulties. We had started by breaking up into shifts, since this was clearly a "hands-on" experiment; there was very little that was automatic about its operation. The accelerator ran continuously, only stopping for occasional source changes or breakdowns. Patrick Somerville spent

long hours at the experiment to assist the students as they learned their roles in watching over everything. Pat's first contact with Seaborg was as a high school student when he attended a lecture that Seaborg gave at the International Science Fair in Baltimore on May 15, 1970. After the lecture he came up to Seaborg and said that he would like to study under him some day and many years later he showed up in Berkeley and did just that, eventually earning his Ph.D. with an excellent thesis in 1984. He obtained the material for his thesis by working tirelessly with both Nitschke and Ghiorso in the SF "wars" with Dubna, so he had an excellent understanding of the physics of the heavy elements. It was largely through his efforts that the puzzle of the identity of $^{260}_{104}$ was solved to everyone's satisfaction. Figure 12.4 shows Pat Somerville and Al Ghiorso in front of SASSY2 just before the element 110 experiment.

Diana Lee tried to keep up with the data by reading the tapes as they were generated in order to detect the occurrence of an interesting event, but the predetermined conditions that were used covered only

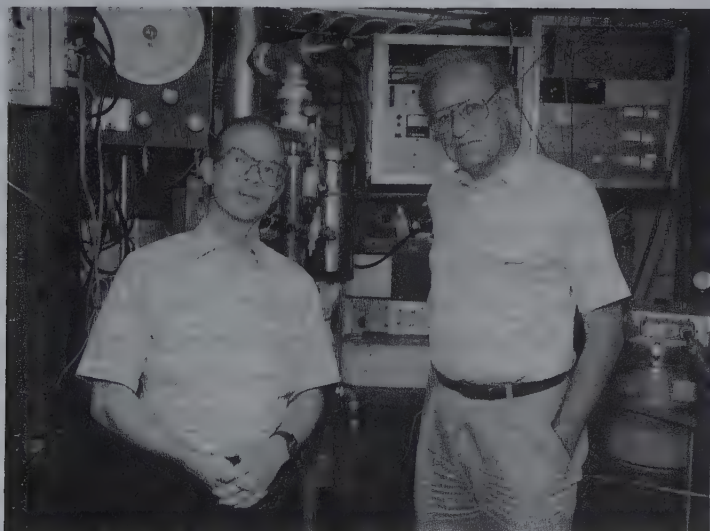


Fig. 12.4. Pat Somerville and Al Ghiorso in front of SASSY2 just before the element 110 experiment.

the obvious scenarios. At the completion of the run, all of the data was re-examined in greater detail by Diana, Phil Wilmarth, and Ghiorso in various ways but still without triggering any of the expected signs that element 110 or its daughters had been produced. It took a lot of time to do a thorough job but that turned out to be highly worthwhile. One of the early runs, RA1017, which had looked interesting at first, was rejected initially because the bombarding energy was thought to be too low and the event characteristics did not fit the profile that had been expected; it was set aside while all of the other runs were being examined. In that process, which took a lot of time, the basic characteristics of SASSY2 both in-beam and out-of-beam were very carefully studied by using the recoil and α -background pulses. By providing examples that could be analyzed and compared and by providing background material, all of the other runs became a laboratory for understanding how SASSY2 functioned. This turned out to be very valuable for unraveling the mysterious event that occurred in RA1017.

Nothing interesting was found in any of the other runs, so we finally returned to this one, now much better prepared to analyze it in detail. The first important finding was that there was excellent internal evidence that the cobalt bombarding energy was much like the others, so there was no reason for ignoring this run. The data showed that an incoming recoil passed through the dE/dX counter with the proper energy loss for element 110 and deposited about 34 MeV while implanting itself a little below the center vertically in crystal 28. This horizontal positon was near the center of the focal plane and thus had the magnetic rigidity that was predicted for element 110. A presumed α -particle with a measured energy of about 10.7 MeV followed 4 μ s after the recoil arrived. When two pulses followed very closely together in time, our studies had shown that the second was decreased in size because of an amplifier effect and that its true energy depended on the time between the two pulses and the height of the first. A double pulse generator was set up to simulate this effect and showed that the true energy of the α -particle was 11.5 MeV, close to the prediction for the α -particle energy for

²⁶⁷110. Walter Loveland also calculated how big the effect would be from the pulse shape and times between pulses and got the same answer. But where was the rest of the α -chain? Approximately at the same position, but 150 ms later, was an out-of-beam α -pulse with an energy of 8.1 MeV, so at first it was assumed that it was a part of the α -energy of the third member of the chain, namely ²⁵⁹106.

The other members of the Coalition were immediately notified by Ghiorso of this exciting new discovery on July 18, 1992, in these words: "Here is a possible scenario. Our candidate comes from 11.5-MeV ²⁶⁷110 decaying 4 μ s after its arrival at the focal plane to its daughter, ²⁶³108, unseen in the 250- μ s 2nd ADC dead-time interval. Following this, the 0.5-s granddaughter, ²⁵⁹106, is observed as an 8.1-MeV alpha rather than 9.03, 9.36, or 9.62 MeV, which are known for this isotope, because it was a partial escape. And following this, the succeeding descendants are presumably lost in the background. An alternate choice might be that the 8.1-MeV alpha is from ²⁵⁵104, but then the 0.15-s decay time becomes harder to justify, though not impossible."

None of us was overwhelmed by this analysis, but it would have to do while we examined it more closely. We soon found out that it was very improbable for a 9-MeV alpha to escape from a crystal and leave only 8.1 MeV. Furthermore, there were a lot of other 8.1-MeV alphas detected out-of-beam in the other runs and these probably came from 25-ms ²¹³Rn kept alive by its electron capture (ec) mother, 35-s ²¹³Fr. Back home again in Oregon, Walter Loveland began to spend a lot of time on the problem analyzing the tapes himself but mainly succeeded only in reinforcing the negative thoughts about the "Candidate." He agreed that the 8.1-MeV alphas could not fit into the scenario and Ghiorso began to tell people that "the Candidate had died of a heart attack." But it did not die after all! About this time in January 1994, Seaborg came up with a brilliant idea and suggested to Ghiorso that the possibility of an ec-mode had been neglected entirely. And he was right! Since this was a highly neutron-deficient region, ec could be a prominent decay mode. At first ²⁵⁵104 was selected as a possibility. This was ruled out for various reasons,

but $^{259}\text{106}$ soon took its place when Ghiorso found an α -particle with an energy of 8.31 MeV at a time 19.7 s after the recoil, which corresponded well to the known 22-s $^{255}\text{103}$.

With this encouragement, a workshop was held at LBL on February 7, 1994, attended by a large number of the Coalition members to debate the merits of the new scenario. Once again we looked at the possibility that the event was accidental and it was agreed that the probability that these events were due to a random bringing together of many individual unassociated events was extremely small, of the order of 10^{-8} or less. After careful consideration it was decided that, on balance, the element 110 interpretation was actually the simplest explanation for the observations. Adding another positive slant, Walt Loveland pointed out that *all* of the seven nuclides above β -stable ^{251}Cf which contain 153 neutrons undergo ec to a significant extent and that for $^{259}\text{106}$ not to do so would be an extraordinary exception. Much later, Victor Ninov pointed out that GSI had measured the ec branch of ^{261}Sg as being 1%, so for ^{259}Sg to have a branch of, say, $\sim 10\%$ does not seem unreasonable.

To summarize this scenario: the fusion product recoil, $^{267}\text{110}$, travels through the helium gas with the proper magnetic rigidity; it loses energy in a proportional counter and deposits energy in the focal plane detector, both amounts being consistent with an atomic number of 110; an α -decay from $^{267}\text{110}$ is observed 4 μs later; the α from $^{263}\text{108}$ is lost during the second ADC dead-time; the $^{259}\text{106}$ ec decay to $^{259}\text{105}$ is not observed; the $^{259}\text{105}$ α -decay is observed only by its escape 2.9 s later; the full energy α -decay of $^{255}\text{103}$ is observed 20 s later; the ec of 4-min $^{251}\text{101}$ and subsequent daughters were not observable in this system. Figure 12.5 shows the proposed decay sequence.

Alternative scenarios, such as might be devised from transfer product decays or random events, were carefully scrutinized and found to be unlikely by orders of magnitude, so we concluded that the association of this event with the formation of element 110 appears to be the simplest and most probable explanation of the observation. Unfortunately, the SuperHILAC was shut down

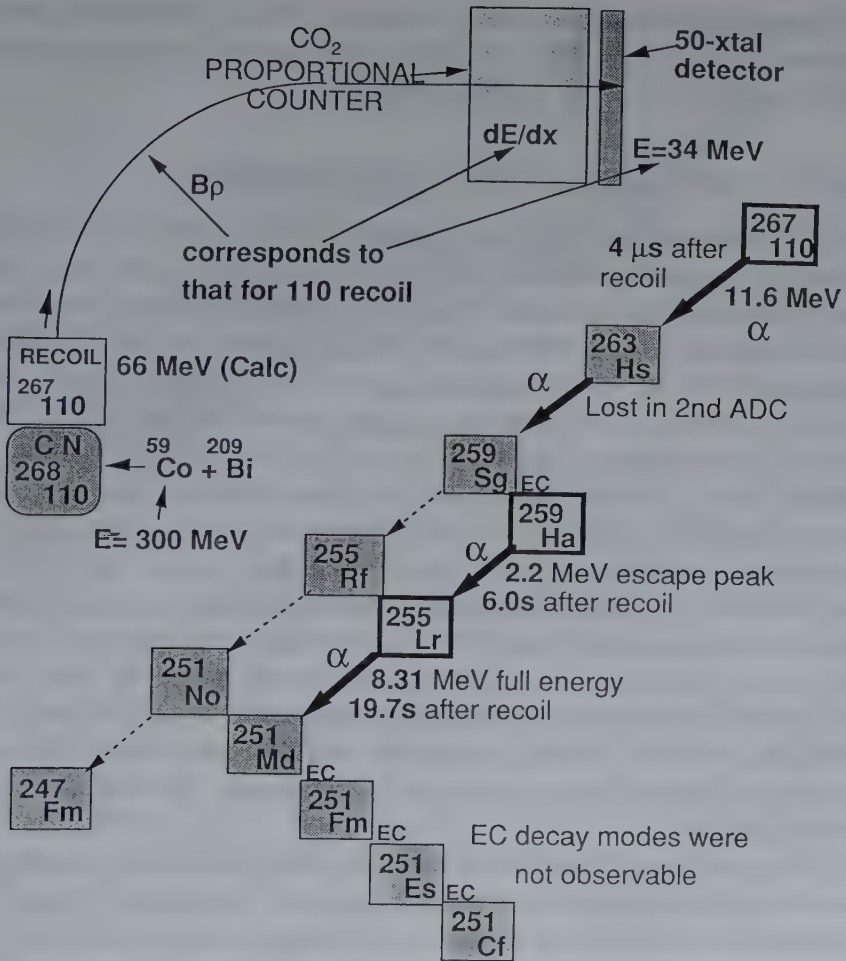


Fig. 12.5. Proposed scenario for the Berkeley element 110 event.

permanently after this experiment was finished and it has not yet been possible to repeat the experiment at another accelerator. Ghiorso first reported^{12,2} on this evidence for $^{267}_{110}$ at the Nucleus-Nucleus Collisions V Conference held in Taormina, Italy, on May 30 to June 4, 1994. A revised version was prepared by Loveland and Ghiorso

and carefully reviewed by the "Coalition." It was published later in *Physical Review C*,^{12,3} after a rather long debate with the referees which resulted in some further revisions.

12.1.2. GSI's Improved SHIP and the UNILAC

At GSI, before undertaking the extremely difficult task of identifying elements beyond element 109, Sigurd Hofmann and his team undertook the task of improving SHIP further. The total acceptance was increased by bringing the target closer to the entrance quadrupoles, and a short dipole to bend the recoil beam 7° was added in front of the detector to give added discrimination. This change, introducing a slight dogleg so that the detector system no longer had a line-of-sight path to the UNILAC, reduced the background by a factor of 10. Most important of all, a new detector system was installed in the form of an almost closed box covered with detecting crystals. An α -particle which escaped through the face of a focal plane crystal now would have a high probability of having the rest of its energy detected by another crystal in the box. With all crystals being position-sensitive, corrections could be made for window thickness effects caused by the emission angle for each α -particle. The optimum size target thickness was determined to be 450 $\mu\text{g}/\text{cm}^2$.

While the improvements to SHIP (Fig. 12.6) were being made, the beam capabilities of the UNILAC were also undergoing important modifications. The basic aim of the changes was to use just one charge state throughout the UNILAC and thus eliminate the stripper losses, which can be very high for the high Z elements. For this purpose a new high-charge-state injector was installed, consisting of a modern ECR ion source on a high voltage platform followed by an RFQ accelerator operating at 108 Mhz. The 27-MHz Wideroe section was bypassed and instead the beam from the RFQ was injected directly into the existing 108-MHz Alvarez linac. With very high efficiency this new arrangement was able to provide steady currents of the very rare (expensive) separated stable isotopes that were

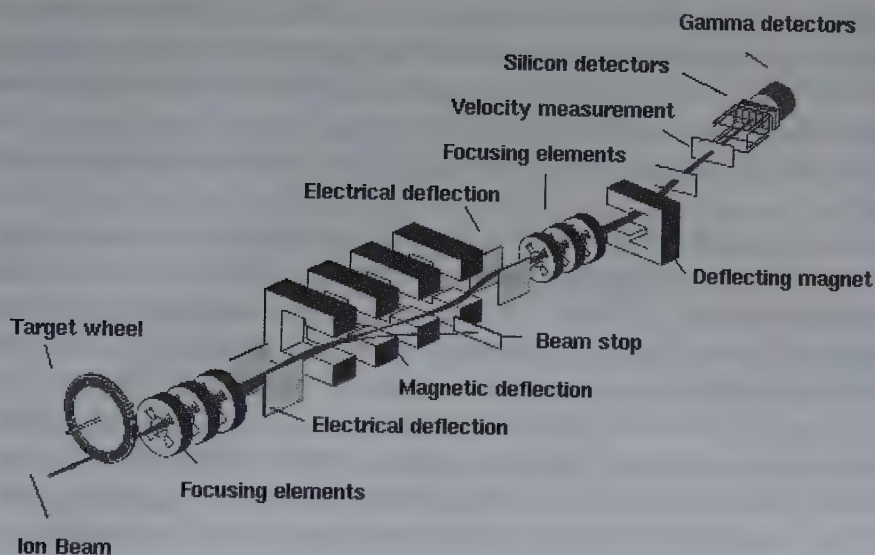


Fig. 12.6. Improvements to SHIP.

needed for the SHIP experiments. Improvements also were made in the UNILAC to increase the operating efficiency in the use of the beam and the ease and accuracy of measuring the beam energy.

These changes paid off handsomely with an estimated 12-fold increase in luminosity, with most of the improvement being due to the UNILAC changes. The new sensitivity (roughly 40% overall) is such that now one heavy-element chain every ten days at a cross section of one pb can be detected using time-averaged beam currents of about 3×10^{12} ions/s. Today the GSI group is recognized as having the premier installation for the study of new elements by "cold fusion" reactions where the recoil distribution is highly collimated in the forward direction. The efficiency of SHIP for reactions like ^{48}Ca on ^{244}Pu is considerably smaller because of the wider angular spread of the fusion recoils.

The spectacular success of the GSI group in uncovering isotopes of elements 110, 111, and 112 within the space of a year is not only

noteworthy in its own right, it is fascinating as an adventure in discovery that deserves to be described in detail. We are indebted to Victor Ninov, one of the participants, for the information that has allowed us to put together the following detailed “inside story” of the trials, tribulations, and triumphs of the Armbruster group in this work.

Even with the much-improved SHIP luminosity the group knew that its next objective, the detection of element 110 in the bombardment of ^{208}Pb by ^{62}Ni particles, would not be easy, because they could only hope to make an atom every week or two. This was made painfully clear by the fact that in the reaction of ^{58}Fe on ^{208}Pb to make $^{265}108$ in the discovery experiments of 1984 the yield at the one energy that they had tried was a meager three events in six weeks. This meant that it was imperative that the proper beam energy be used, since the excitation functions for similar reactions in this region had been shown to be very narrow; an energy wrong by a few MeV was enough to drop the yield by a factor of 3 or more.

Was there a way to predict the proper energy to use? It was decided that their best strategy was to accurately determine the excitation functions for the lighter even- Z elements, 104, 106, and 108, and then extrapolate up to 110. From earlier results with reactions that made element 104 they had been intrigued by the possibility that the optimal energy needed might be *below* the so-called Bass Coulomb barrier, so they decided that since they already had a fairly good yield curve for 106 they would first go back and remeasure the 104-excitation function more accurately. If they did the same thing for 108 they would then have three energy points, which should allow them to extrapolate up to 110 with much more confidence.

In the summer of 1994 they started their 104-excitation function and in a few weeks had a precise curve. The next step was element 108. They started with the ^{58}Fe energy previously used in 1984 (5.019 MeV/N) and ran for one week without getting a single event. Panic — was everything set properly? Statistics can play strange tricks — don’t touch anything! Finally, on Friday, they got two events — one alpha and one fission delayed by 150 μs ! The α -particle

decayed into the most beautiful decay chain that they had ever seen because of the new detector box, the implantation plus four- α -decay-chain going all the way down through ^{253}No .

But what about the fission? Was it SF branching of $^{265}108$ or was it $^{264}108$ from the 2-n out reaction? So they transferred that energy point into the excitation plot that they had just made for the production of isotopes of element 104 — and found that the equivalent 108-point was at the crossing of the 1-n and 2-n excitation functions! Almost surely that fission must be from the 2-n reaction to make $^{264}108$ (later work showed that this isotope had a 50% SF branch). Now, should they go up in energy or down? At this point there was a divergence of opinion as to what the optimum bombarding energy should be to make $^{265}108$. Armbruster thought that they should use a higher energy than had been used in the original experiment in 1984, on the reasonable basis that the theorists had calculated that an “extra push,” i.e., an energy slightly above the classical barrier calculation, would be needed to get the heavy projectile to fuse with the target. The younger members of the group felt that a lower energy should be used. What should they do? The element 108 events had come in very slowly in the original experiment, something like one event every two weeks, and supposedly they were already at the optimum energy. If they used the wrong energy the rate would be even lower. They debated at length all of the scientific arguments that they were aware of, but could not come to a clear consensus. Finally, someone proposed that they use the democratic solution — voting!

Unprecedented! And they voted to go down in energy, with only Peter Armbruster dissenting. He exploded and said that it was the first time in the history of science that the direction of an experiment would be decided by a democratic process and went away muttering, “If you introduce democracy into physics you can only make mistakes!” But it was not a mistake. He went away for half a day. Of course, he calmed down, and the next day everything was fine, because it was already clear that the cross section had increased — in one day they had a couple of events! They proceeded to make the

energy intervals small, about 0.1 MeV/N. The question then was, How high would the cross section go?

They drew a Gaussian curve where they thought it should be on the basis of the 104 and 106 data and each member of the team guessed where the points would fall and placed initials to correspond. Oganessian happened to be there on that day and added his guess, too. The prize was 1 mark.

They decided to run long enough to get at least three events at each point — they got some 15 events in about 2 days at the peak of the excitation curve. Of course, Peter was very happy when the cross section went up so spectacularly, but he wanted to get on with the 110 experiment itself by switching over to ^{62}Ni immediately. He knew that the Dubna–Livermore experiment at Dubna to make 110 by the $^{244}\text{Pu}(^{36}\text{S},\text{xn})$ reaction was also running and he was worrying that they might be scooped. It is understandable that his sense of urgency caused him to be somewhat difficult from time to time about the delay. Peter would come into the office as they were reducing the data and drawing beautiful α -decay chains which looked like stamps, and say things like “We are here to discover heavy elements, not collect stamps!”

This single-minded fixation on wanting to quickly move on to the ultimate goal rather than taking time to do a thorough and publishable job on the more “pedestrian,” although important, preparatory experiments, is not unique to Armbruster. It seems to be a characteristic of many visionary and charismatic scientific leaders.

On the other hand, the team wanted to get good data so that the energy to make 110 could be predicted more accurately, and thought that the time to get a good excitation curve for 108 would be well spent. They too were in a hurry to switch over to nickel beams but this was their “insurance” and they wanted to do the excitation function for 108 as well as possible, so again they insisted on doing it their way. It was a tribute to the rapport between Armbruster and his group that they could argue so effectively with the one who was the unquestioned brilliant leader who had managed to bring them to

their position of unchallenged world leadership in the search for new elements.

This was in October 1994. In November, when they did switch over to ^{62}Ni , the functions had been done so accurately that extrapolating with a straight line from the peak energy to make 104 through the peak energy to make 108 gave exactly the right energy to make 110, so their time was indeed well spent. In the first three days of bombardment of ^{208}Pb by ^{62}Ni they had three events of 110! After they had detected the first $^{269}110$ α -chain in the late afternoon of November 9, 1996, they decided that the next step was to write the paper, so this they proceeded to do. The major paper writers, Sigurd, Fritz Hessberger, and Victor, disappeared into their offices to start this enjoyable task, leaving word that they did not want to be disturbed, so at this stage they did not tell the many guests at the UNILAC who had heard that they were trying to make element 110 that the discovery had already been made. Even Peter was not told, because they wanted to present him with an elegant surprise. Drafting the essential ingredients for the manuscript took a few hours but finally they finished it about 3 a.m. and left it on Peter's desk with a note that now it was his turn and that they would not be in until noon. Of course, he was extremely happy and made his comments and corrections. There were another two events by this time. They got one more event in the next eight days. These made beautiful "stamps."

At this point they debated a little about whether they should switch to ^{64}Ni or go on to 111, and decided that since they had a lot of time left, about six weeks, they should finish element 110 first so that they did not have to come back to it. That was the right thing to do, because the cross section was even larger to make $^{271}110$ than it had been to produce $^{269}110$. They were actually able to measure a good excitation function for 110, something that had not even been dreamed of before. Altogether, they had found two isotopes of 110, four events of one mass and some fifteen of the other mass at three different energies. At this point they shipped their paper off to Heidelberg to *Zeitschrift für Physik A* by special courier, and in

November 1994 announced their discoveries of the two new isotopes of 110 to the world.^{12.4} The characteristics of the two were: $^{269}\text{110}$ (170-ms, 11.13-MeV α), produced in the reaction $^{208}\text{Pb}(^{62}\text{Ni},n)^{269}\text{110}$, the identification being made by the observation of four correlated α -decay chains; and $^{271}\text{110}$ (1.4-ms, 10.68-MeV α), produced with an even larger cross section by using ^{64}Ni on ^{208}Pb and identified by seven correlated α -decay chains. Their detection system worked so well that they were able to program their computer to reduce the data on-line so that each event in a chain was labeled according to Z and A! They joked that they might just as well have programmed the computer to write the paper. This magnificent accomplishment meant that they had entered a new regime in which they could confirm their own discoveries by producing enough α -decay chains to have unambiguous proof of their findings.

They knew about the possible $^{267}\text{110}$ event which had been reported by Ghiorso several months before at the Conference in Taormina, Sicily, but did not care; nor did they not care about the Dubna–Livermore experiment which claimed a possible $^{273}\text{110}$ event. They simply wanted to do the best possible identification so that there was no doubt about *their* work.

12.1.3. Dubna's Gas-Filled Separator

The Dubna–Livermore experiment on element 110 ran simultaneously with the GSI work and their result was announced^{12.5} in January 1995. Members of a Russian–American collaboration led by Yuri A. Lazarev* of Dubna and Ronald W. Lougheed of the Lawrence Livermore National Laboratory used the Dubna small angle, gas-filled separator. This device is very similar to the original Berkeley SASSY except that it uses both a rotating target wheel and a rotating window wheel so that large accelerator currents can be used to counteract the rather low efficiency of the separator. After a long experiment, the experimenters claimed the observation

*Deceased April 1996.

of one atom of still another isotope of element 110 in the reaction $^{244}\text{Pu}(^{34}\text{S},5\text{n})^{273}110$. It also was identified by the observation of its α -emitting descendants. It should be noted especially that this is a "hot" fusion reaction with five neutrons emitted! Apparently, proximity to the predicted deformed subshell at 162 neutrons led to the relatively long half-life of 0.3 ms for this isotope which decays by emission of 11.4-MeV α -particles. However, the background was far from being zero and it is possible that the chain of α -events that were used for the assignment of one of the events to $^{273}110$ could be due to random events. A later paper published by the Dubna-Livermore group^{12,6} had a list of 18 possible events, from which the final chain was chosen. This circumstance substantially reduces one's confidence that it indeed was due to element 110. However, just as for the single Berkeley event, final judgement on this claim should be withheld pending its confirmation.

12.2. Element 111

Now it was time to go for 111. To do this the GSI team had merely to change from the ^{208}Pb target to ^{209}Bi . Everything else stayed the same, including the energy that they had used for 110. But it wasn't as easy as with the even-Z elements. They saw nothing at first, because the energy was too low. The odd-Z elements need a little more energy. It turned out that they had good excitation function data for 107 but not for 105 or 103, and they had only the one energy point for 109. After a few days with no events they had to change the ion source, so Victor with the acquiescence of Sigurd decided to increase the energy simultaneously so as not to waste any time. It usually took about the same amount of time to change the source as to change the energy, so they tried to synchronize those two chores as a matter of policy.

This was in the evening. The next morning, at 9 a.m., when Peter came in he examined the Log Book and found that they had changed the energy. He was quite upset at first, until Victor calmed him down by saying that Sigurd and Peter were naturally preoccupied

with dealing with the Press about the element 110 discoveries at the same time that there was a running experiment to attend to. Fritz and he had carefully discussed the energy change, and Sigurd had told them to do what they thought best. And it *was* the correct move. There were no counts at the lowest energy, one at the intermediate energy and then two at the highest energy, so they still didn't know where the peak was. At this stage they did not care, because they had won a clear title to the discovery of element 111 and had set the stage for going beyond! As luck would have it, only one of the $^{272}_{111}$ alphas in the three chains was totally absorbed in the detector system, giving an energy of 10.82 MeV. The three events, showing a half-life of 1.5 ms, led directly through the unknown nuclides, $^{268}_{109}$ and $^{264}_{107}$, to $^{260}_{\text{Ha}}$, the same nucleus that had been discovered 24 years earlier in Berkeley.

A short note was sent off to *Zeitschrift für Physik A* on December 12, 1994: "The New Element 111," by Hofmann, Ninov, Hessberger, Armbruster, Folger, Münzenberg, Schott, Popeko, Yereimin, Andreyev, Saro, Janik, and Leino.^{12.7}

At this point they stopped for the Christmas shutdown, completely exhausted, and went on holiday. They essentially had been running a continuous experiment from June until the end of the year! They did not run on shifts because they had automated SHIP so well that it usually ran unattended, with someone always on call. It was necessary to replace the target wheels every three to five days because of the damage caused by the 1.2 pμA of peak beam current and there were various other routines, such as changing the energy and the tapes, checking on the monitoring devices, calibrations, etc. These jobs were handled by everyone, but the core group making the big decisions — Sigurd Hofmann, Victor Ninov, Fritz Hessburger, Gottfried Münzenberg, Matti Leino, and Peter Armbruster — spent long hours constantly hovering over the experiment every day and night.

When they came back after the refreshing holidays Sigurd began promoting an interesting idea. He reasoned that it was possible for the interaction barrier to be substantially lowered by the preliminary

melding of a proton pair as the two particles came close together and thus it might be possible to form element 116 with a 0-n reaction. Why not bombard ^{208}Pb with ^{82}Se ? It certainly seemed like a reasonable idea and if it succeeded they would not only make element 116, they would also produce elements 114 and 112 as alpha decay daughters! They made many calculations of the needed energy using various mass formulas and assumptions as to nuclear radii, but it seemed that there were a lot of uncertainties and they ended up using six different energies. They ran for six weeks without seeing anything down to the 3-picobarn level. It was a good try, with everyone hopeful but not convinced that they would be as lucky as they had been with 110 and 111.

12.3. Element 112

So it was back to more familiar ground and they began to bombard the familiar ^{208}Pb targets, this time with ^{70}Zn to make element 112. They ran in January 1996 for six weeks, their enthusiasm significantly lowered compared to the 108 and 110 experiments. (Ninov said that it should not be surprising that there should be a letdown because it had been at such a high level before and everyone was very tired from their prolonged "big push.") For some unknown reason the sources behaved erratically. The first source performed beautifully for four weeks giving them their usual 600-pnA average current, but the second had problems. They ran only at one energy and with a 1-pb cross section got two events with the unmistakable signature of the nuclide chain starting with $^{277}112$. Two very long α -sequences were observed, one with five members and the other with six, both passing through 78-s ^{261}Rf , one of the isotopes of rutherfordium discovered at Berkeley three decades before. Two slightly different α -energies were measured, 11.65 and 11.45 MeV for the 112 decay with an average $T_{1/2}$ of 240 ms. This work was published on February 21, 1996, in *Zeitschrift für Physik A* as a short note: "The New Element 112," by Hofmann *et al.*^{12,8} with the same authors as for the element 111 paper. The group is shown in Fig. 12.7.



Fig. 12.7. *Front row, left to right: V. Ninov, S. Hofmann, P. Armbruster, S. Saro, A. Yereimin, and S. Raiss. Back row, left to right: F.-P. Hessberger, G. Münzenberg, A. Popeko, H. Folger, H.-J. Schott, C. Stodel, and H.-G. Burkhard.*

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Chapter 13

Naming Controversies and the Transfermium Working Group

13.1. Establishment of the Transfermium Working Group

The 1974 international *ad hoc* committee which was established to consider the claims of priority of the discovery of elements 104 and 105 and to try to achieve some agreement between the laboratories at Berkeley (USA) and Dubna (USSR) by exchanging representatives between the laboratories and encouraging them to resolve their differences was finally disbanded in 1984. Although the committee never met as such, it had facilitated meetings between the scientists involved, but to no avail, and no resolution of the conflicting views resulted. The Chairman, Prof. J. Lewis, did request that both Dubna and Berkeley prepare documents outlining all of their work and their publications on elements 104 and 105. That was done by the American members, as discussed in detail in Chapter 9. However, essentially no progress was made concerning official recognition of the priority of discovery or of the names for these elements, and each group continued to use the names they had originally proposed — rutherfordium (Rf) and hahnium (Ha) by Berkeley, and kurchatovium (Ku) and nielsbohrium (Ns) by Dubna.

After the reported discoveries of elements 107, 108, and 109 at GSI between 1981 and 1984, Peter Armbruster, the leader of the GSI team, published an article^{13.1} in 1985 entitled "On the Production of Heavy Elements by Cold Fusion: The Elements 106 to 109," in which he proposed a "rule" (see paragraph reproduced in Fig. 13.1) to be applied retrospectively for all elements discovered by *isotope*

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ON THE PRODUCTION OF HEAVY ELEMENTS BY COLD FUSION: The Elements 106 to 109

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4.5 The Naming of Elements

Not hiding a certain self-interest, we want to help settle existing controversies on the naming of elements (103, 104). As a rule we propose the following: Element synthesis becomes production of a given isotopes, and a name should be accepted only if the experiment claiming the discovery is reproducible. An isotope is define by its mass and atomic number, its fingerprint are its decay modes and its half-life. Decay modes of the heavy isotopes in question are electrons capture and spontaneous fission, which sometimes are difficult to assign to a specific isotope, and a decay, which (with its decay energies measured with an accuracy of some parts in a thousand) gives a very reliable mode of assignment. The time correlation of subsequent decays is a further method of definite isotope identification. The proposed rule should be applied retrospectively for all elements discovered by isotope identification, that is elements 102-109. A joint commission of physicists and nuclear chemist urgently is needed to solve that long-standing controversies

Fig. 13.1. Paragraph from Ref. 13.1, Section 4.5, p. 175. "On the Production of Heavy Elements by Cold Fusion: The Elements 106 to 109," by Peter Armbruster.

identification, i.e., elements 102–109. He further suggested that a joint commission of physicists and nuclear chemists was needed to solve the longstanding controversies surrounding the naming of these elements. Perhaps in response to his urging, the IUPAP subsequently decided it was time to appoint a Transfermium Working Group (TWG) to look into the priority of the discovery of all the elements heavier than fermium ($Z=100$) so that official names could

be proposed. And, in 1986, D. Allan Bromley, President of the IUPAP (1984–1987), proceeded with the appointment of a TWG. Seaborg was asked to suggest names for the TWG and he suggested six internationally known, “neutral” nuclear chemists, but none of these was selected. The IUPAC also asked that it be represented, since naming of the elements clearly was within their historical purview. The Executive Boards of the IUPAP and the IUPAC agreed to the following two phases: (1) the establishment of criteria that must be satisfied for the discovery of new chemical elements to be recognized; (2) the application of those criteria in practice.

By May 1987 the IUPAP/IUPAC Working Group on Transfermium Elements had been established, with Sir Denys Wilkinson, University of Sussex, Brighton, UK, as Chairman and the following membership: R.C. Barber, Winnipeg, Canada; N.N. Greenwood, Leeds, UK; A.Z. Hryniewicz, Krakow, Poland; Y.P. Jeannin, Paris, France; M. Lefort, Orsay, France; M. Sakai, Tokyo, Japan; I. Ulehla, Prague, Czechoslovakia; A.H. Wapstra, Amsterdam, The Netherlands. One of the problems with the TWG was that although all the members were eminent scientists, there were no heavy element nuclear chemists or radiochemists, nor indeed any radiochemists, amongst its membership. Furthermore, only two (Greenwood and Jeannin) of the nine members were representatives of the IUPAC. Originally it was envisioned that this group would only define the criteria to be used in determining whether or not a new element had been discovered and that they should work iteratively with the major experimental laboratories concerned to define criteria that would command general assent, before consideration was given to Phase 2.

13.2. TWG Visit to Berkeley

Although the TWG visited Berkeley from June 19 to 21, 1989,^{13.2, 13.3} they seemed uninterested in the complete history and bibliography of the work of all laboratories on elements 104 and 105 prepared by Hyde, Hoffman, and Keller.^{9.1} Copies of this document were given to Wladek Swiatecki of LBL, who met with the TWG on the first day

(Monday, June 19), but they made no attempt to meet privately with either Hyde or Hoffman, who were both at LBL at the time. The next day the nine members of the TWG met with Ghiorso, Hoffman, Matti Nurmia, Seaborg, and Pat Somerville from 9 a.m. to about noon in the LBL Director's Conference Room. (See Fig. 13.2 for photo of group.) Sir Denys Wilkinson, TWG Chairman, made some introductory remarks indicating that they were only concerned with Phase 1, i.e., outlining the criteria to be used in determining whether a new element had been discovered, and that they wished to develop these interactively with the laboratories involved. He hoped to report on Phase 1 by Fall 1990. However, he did say they would inevitably be concerned with some of the details of discovery, but emphasized that neither Phase 1 nor Phase 2 would be concerned with the actual names to be assigned to the elements.



Fig. 13.2. Photo from Ref. 13.2, p. 247f. Visit of the IUPAP/IUPAC Transfermium Working Group to the Lawrence Berkeley Laboratory, June 20, 1989. Front row, left to right: I. Ulehla, Sir Denys Wilkinson (Chairman), Glenn T. Seaborg, and Y. Jeannin. Back row, left to right: M. Lefort, N.N. Greenwood, A. Hryniewicz, M. Sakai, R.C. Barber, and A.H. Wapstra.

Seaborg discussed the paper "Criteria for the Discovery of Chemical Elements" published in 1976^{9,3} by an international group whose authors also included Hoffman, Hyde, Keller, and Lefort, a member of the TWG. He emphasized the view expressed in that paper that the atomic number should be determined by some method such as the genetic alpha decay sequence or measurement of the characteristic X-rays. He reminded the committee that the names "mendelevium," "nobelium," and "lawrencium" for elements 101, 102, and 103 had already been approved by the IUPAC Committee on the Nomenclature of Inorganic Chemistry (CNIC) in 1970 and that new criteria for discovery should not be retroactively applied to those elements. [The Commission on the Nomenclature of Inorganic Chemistry (CNIC) published "Nomenclature of Inorganic Chemistry," 2nd Edition, "Definitive Rules 1970." See Ref. 13.4, p. 98, in which Table I (Elements) lists element 101 as mendelevium, Md; element 102 as nobelium, No; and element 103 as lawrencium, Lr.]

An earlier "Criteria" paper had been published in 1971 by Flerov and Zvara of the Dubna group,^{9,4} which also expressed the viewpoint that the claim to discovery of a new element should be related closely to its atomic number and that "if the atomic number is found chemically or through the atomic physical techniques (x-ray spectroscopy, etc.), then the work will be (correspond to) the discovery even if no nuclear physical identification is used. In this case, the mass number is not necessary to be found at all." However, they also stated that "when radioactive properties alone of an element are studied while the identification is nuclear-physical, the discovery will be real only if the conclusions on both the mass number and the atomic number have not been revised later." They further stated, "The atomic number of the element should be found unambiguously while the decay properties of the isotopes (should be) measured accurately to the level of up-to-date experimental techniques. The mass number should be proved to be unambiguous or else the uncertainty, if present, should be discussed explicitly."

Wilkinson said that he agreed with Seaborg and Hoffman about the primary criteria, but that the Dubna group believed in the

efficacy of reaction mechanisms. A rather wide-ranging discussion followed, with Hoffman pointing out the pitfalls in using spontaneous fission alone as a method of determining atomic number based on the half-life or the excitation functions for the assumed compound nucleus reaction. The SF experimental half-lives show a wide range of values which overlap the various elements. In addition, recent measurements by Hoffman's group of systems, such as ^{248}Cm with ^{18}O and ^{22}Ne projectiles, showed that significant yields of nearly all the elements between the target Z and that of the compound nucleus can be produced. There was a discussion of the 1969 gas phase chemical experiments of Zvara and coworkers in which they reportedly showed that the originally claimed 300-ms SF isotope formed in the $^{242}\text{Pu}(^{22}\text{Ne}, 4n)^{260}104$ reaction behaved in volatility experiments like the group 4 element hafnium and, therefore, must be 104. (See our detailed discussion of this in Chapter 9.) The Berkeley group pointed out that Zvara had said they could only determine the half-life if it had a value in the range of the transit time across the mica detectors which was about 0.7 s and they found that the distribution of fissions in the mica detectors was consistent with a half-life of 0.3 s. They then concluded that the fission track distribution "shows that the effect was not caused to an appreciable extent by the decay of nuclides undergoing spontaneous fission with half-lives of 0.014 s and 3.7 s." (The two prominent SF half-lives observed in their original decay curves were 0.014 s from $^{242}\text{Am}^m$ and 3.7 s from ^{256}No .) Thus it seemed clear that they could not have separated the now known 20-millisecond SF activity of $^{260}104$, because it would have decayed before reaching their detectors. Jeannin asked if Zvara's group shouldn't still see the short-lived activity if it went through their columns, but Hoffman reiterated that the transport time was too long. Wapstra said that Flerov wanted to "use the 3-second isotope," but both Hoffman and Seaborg reiterated that Zvara had disclaimed that half-life. Hoffman added that thermochromatography of the transactinides was not so well known and that her group and others were still trying to understand it better using *isotopes of known elements*. Jeannin added^{13.3} that "they

(Zvara *et al.*) claim that 104 is eka-hafnium and 105 is eka-tantalum but their peak for 105 is at hafnium!"

Ghiorso pointed out that the Berkeley group had used SF in the discovery of element 101 but that chemistry was also used and that reaction mechanisms do not help. Lefort asked about the use of excitation functions and angular distributions — Ghiorso and Seaborg both said it could not be done with sufficient accuracy.

Seaborg made the suggestion^{13,2} that the TWG should continue into Phase 2, which was to be the actual application of the discovery criteria to determine who the discoverers were, and that if the Berkeley discoveries of elements 104, 105, and 106 were confirmed, the name "kurchatovium" might be suggested for element 106. In retrospect, Hoffman thinks this is one of Seaborg's very few ill-advised suggestions!

There was some discussion of the need for confirmation of a discovery before the discovery group proposed a name. Wilkinson said that perhaps the IUPAC might relax the requirement for confirmation as a condition for naming. Hoffman said she didn't like such a relaxation and that honoring this requirement in the case of 106 avoided the conflicts which had been associated with 104 and 105. Seaborg again said that division of the work into Phases 1 and 2 was not realistic and that this TWG should do it all! Wilkinson asked if a different symbol for kurchatovium, such as Kt, could be given to element 106, and said, "I take it that you do not like names such as 'unnilquadium.'" There was a quick response of "No!!" from the Berkeley scientists.

Ghiorso offered that his group could confirm 107, thus also helping to confirm the GSI discovery of element 107. Hoffman suggested that if the TWG would say that it was important to confirm 106, it would help her group get Cyclotron time to perform such confirmation experiments, which they were equipped to do. However, Jeannin said that the Committee on Nomenclature never asked for confirmation experiments, but Seaborg reminded the group that the "Criteria" paper of 1976^{9,3} suggested the need for confirmation and that this would avoid a repetition of the 104 and 105 controversy.

Hoffman said that in practice the Russians had also agreed that discoveries should be confirmed before names were proposed.

That evening the TWG members, including Mrs. Wilkinson and Mrs. Wapstra, together with Al Ghiorso, Darleane Hoffman, John and Louise Rasmussen, Helen and Glenn Seaborg, and Wladek and Mrs. Swiatecki, had dinner in the Lewis-Latimer Room at the Faculty Club on the University of California, Berkeley, Campus. Seaborg made some informal remarks about Berkeley 50 years ago and Gilbert N. Lewis and Wendell M. Latimer, for whom the room was named, and others contributed stories and reminiscences. It was a very cordial evening and there was general discussion among the participants.

The TWG met again on Wednesday morning, June 21, 1989, with Ghiorso, Hoffman, Nurmia, Seaborg, Somerville, Jose Alonso, and Michael Nitschke (from Berkeley), and Kenneth Hulet and Ronald Loughheed (both from Livermore and members of the element 106 discovery team). The discussion on Criteria continued with Wapstra asking a number of questions about details of the Berkeley work on elements 102, 103, and 104. Hryniewicz asked several questions about Zvara's work on trying to identify $^{260}104$ with a 0.3-s half-life by volatilization of the tetrachloride and both Hoffman and Seaborg again pointed out that his work was inconsistent with a 20-ms half-life for this isotope and that Zvara himself claimed that his experiments were inconsistent with the 3-s half-life of $^{259}104$.

There was some more discussion about Criteria and Nurmia^{13.3} stated that there were two basic differences between the Criteria paper of Harvey *et al.*^{9.3} and Flerov and Zvara.^{9.4} "The first was published as the consensus of an international group and the main point is the unequivocal establishment of Z. Furthermore, none of the authors was involved in the dispute. The second paper represents only the Dubna view and the main point is that the ("reported") results are not subject to revision. This encourages publication of poor results in the hope that one is right."

There was additional discussion about having an international tribunal to review claims, and various ideas were discussed. Nitschke

proposed just having dual names for awhile and waiting and seeing who was correct. Hoffman suggested having a list of worthy names which could just be assigned in turn as the elements were discovered, thus depoliticizing the process. But Seaborg cautioned that naming of elements by the discoverers had a long history and that current discoverers would not want to relinquish this right.

The discussion then turned to what constitutes an acceptable means of publication. Various TWG members expressed opinions concerning whether unpublished laboratory reports — such as JINR or LBL reports, which have limited distribution — were acceptable. Hulet said he had serious problems trying to get the Dubna JINR reports in the libraries and even by direct request to the authors. Several present expressed the opinion that only refereed journals were acceptable for either discovery or confirmation reports, but Prof. Sakai observed, "Things are not equally open in the Socialist countries." Wapstra averred, "We need to go to Dubna with an informed but open mind. You take a hard line about publications." Seaborg responded, "Because all other lines are so murky!" The meeting then adjourned for lunch.

After a 2 p.m. seminar by Prof. Sakai, Seaborg met with the nine TWG members in his office. Among other topics, the motive for Dubna's delay of the TWG visit there from April of 1989 to February of 1990 was discussed. Seaborg proposed that it was because: "(1) they probably would like to see the visit not take place and, therefore, a long postponement might lead to something like that, and (2) they probably wanted to have the last word — i.e., be the last place visited."

Jeannin raised the question of whether he should continue on the TWG since he was incoming president of the IUPAC, but it was later decided that it was best for him to continue until Phase 1 was completed, but resign from the continuing committee, which would consider Phase 2.

Seaborg notes that later that evening he read part of the Dubna paper furnished to the TWG and that it was devoted only to elements 101 through 103 and was very long.

The TWG subsequently visited Flerov's group at Dubna, USSR. After the TWG meeting at Berkeley there was a long interval, with no further formal communication between them and the Berkeley or Livermore groups. Nor were there any further requests for information or clarification, nor any chance to comment on any information furnished by the Dubna group during the TWG visit there. No comment was requested on any preliminary views on criteria or ideas on assignment of credit for the discovery of the transfermium elements. Nor was any opportunity afforded the US groups to comment on the accuracy of any draft reports from the TWG.

However, just after the 1990 Welch Conference in Houston, Texas, on "Fifty Years with Transuranium Elements," Yuri Oganessian came to visit at Berkeley and on October 24, 1990, met with Ghiorso, Seaborg, and Earl Hyde, in Seaborg's office,^{13.5} to discuss a possible compromise on the naming of elements 102–106. Seaborg suggested that the names "nobelium" for 102 and "lawrencium" for 103 be kept, but that Berkeley would be willing to share credit for these discoveries with the Dubna scientists. He further proposed that "rutherfordium" be kept for element 104, because the Dubna evidence did not support their right to share the discovery, and that "hahnium" be retained for 105, but that the contribution of the Dubna scientists was large enough that they should perhaps share the 105 discovery with the LBL scientists. Seaborg went on to say that credit for the discovery of element 106 should accrue to the LBL scientists, but that nevertheless he would be willing to recommend that it be named "kurchatovium." Oganessian strongly supported the name "kurchatovium" for element 104 and Hyde and Ghiorso (although preferring "rutherfordium") seemed willing to concede this. The meeting resumed the next afternoon, and this time Darleane Hoffman joined the group. In earlier conversations with Seaborg she had expressed agreement about "kurchatovium" for element 106 and was adamant about keeping "rutherfordium" at 104, and tried to convince Earl and Al of this. Oganessian implied that he would take the naming suggestions back to Flerov. He also reiterated an

invitation to Ken Czerwinski (a graduate student of Darleane's working on rutherfordium chemistry) to come to Dubna to work with his group and that he would send someone to Berkeley to work with Al. We were all rather encouraged by these discussions.

Again, on August 27, 1991, during the American Chemical Society meeting in New York City, Seaborg reiterated to Oganessian that a compromise along the following lines might be possible. The Berkeley group would be willing to agree to codiscovery with Dubna of elements 102, 103, and 105, keeping the names "nobelium," "lawrencium," and "hahnium," while the Dubna group would acknowledge the Berkeley claim to the discovery of element 104 and that the name "kurchatovium" would be suggested for element 106. Seaborg observed, "Oganessian did not appear to want to agree to this."^{13.6} In retrospect, this may have indicated his optimism, or perhaps even knowledge, about the forthcoming Phase 2 report of the TWG, which had just been approved for publication by the IUPAC Bureau in its August 1991 meeting in Hamburg, Germany, but which was not accepted by the IUPAP Council until September 1991, in Madrid.

13.3. Assignment of Priority of Discovery for Elements 101 Through 109

The TWG Phase 1 report was accepted in 1990 by both the IUPAP and the IUPAC and published in *Pure and Applied Chemistry*^{13.7} in late 1991. As indicated, the report of Phase 2, the judgmental phase of the work of the TWG, was quickly approved for publication by the IUPAC Bureau in its meeting in Hamburg in August 1991 and accepted by the IUPAP Council at its September 1991 meeting in Madrid. These precipitate actions came like a "thunderbolt" and were a complete surprise to the Berkeley group! They had understood that according to the original concept, the TWG would work "iteratively" with the major laboratories involved and that there would be a chance to discuss and comment on the findings before they were summarily published. Seaborg obtained copies of the

Table 13.1. 1992 TWG Assignment of Credit for Discovery of Elements 101–107.

Element	Discoverers
101	Berkeley
102	Dubna
103	(Confidence built up over a decade) Berkeley: 1961, 1971; Dubna: 1965–70
104	Shared: Berkeley/Dubna
105	Shared: Berkeley/Dubna
106	LBL/LLNL team
107	Shared: GSI/Dubna
108	GSI
109	GSI

reports to be published and the TWG assignments for priority of discovery are shown in Table 13.1.

The TWG essentially reinterpreted some of the early Dubna gas phase chemistry work on element 104 and decided it constituted proof that element 104 had been identified even though the gas phase chemistry was too slow to permit isolation and detection of the 20-ms SF activity which was subsequently assigned to $^{260}\text{104}$. In summary, the 1964 original Dubna claim to priority of the discovery of element 104 was based on physical detection of a 300-ms SF isotope. The Dubna group later shortened the half-life to 80 ms, and much later they finally assigned a 20-ms SF activity to $^{260}\text{104}$. Obviously, the gas phase chemical separation was too slow to constitute proof of the discovery of an isotope as short as 20 ms. So the TWG said instead that Zvara *et al.* had detected SF decay from the 3-s $^{259}\text{104}$, identified in 1969 by Ghiorso *et al.*^{9,2} based on its α -decay. This was in spite of the fact that in the original 1969 Dubna report^{9,15} Zvara *et al.* stated that the fission track distribution “shows positively that the effect was not caused to an appreciable extent

by the decay of nuclides undergoing spontaneous fission with half-lives of 0.014 s and 3.7 s. Furthermore, the SF branch of $^{259}\text{104}$ was reported to be only of the order of 6% by Bemis *et al.*^{13,8} in 1981 and several more recent attempts to measure it have resulted in even lower limits. This means that Zvara *et al.*^{9,15} could not have been detecting even the 3-s $^{259}\text{104}$, and certainly not the now known 20-ms $^{260}\text{104}$, thus effectively destroying the Dubna claim to priority of the discovery of element 104. Nevertheless, the TWG still said that Dubna should share the credit for the discovery of element 104 with the Berkeley group!

The biased and many erroneous conclusions based on "reinterpretations" of the Dubna data, especially on element 104, and the arbitrary decision to review elements 101 through 103, and the "downgrading of the discovery of element 101 in 1955 by the Berkeley Group" were greeted with disbelief and outrage at Berkeley. Seaborg and Ghiorso quickly prepared a brief (two-page) response^{13,9} which was sent via fax on November 19, 1991, to Wapstra, secretary of the TWG. In it they expressed their "keen sense of disappointment... on what we consider a seriously flawed report, 'Discovery of the Transfermium Elements.' ...The report is so deficient in content that it is difficult to prepare a short comment." The complete text of this short letter, which reiterates many of the points on 104 discussed in this section as well as commenting on the TWG treatment of 101, 106, and 108, is given in Fig. 13.3 and speaks for itself. An additional letter reiterating and expanding on the element 104 situation was sent on November 26, 1991.^{13,10} The response from the Berkeley group attempted to point out these and other failings in the TWG reports and that the flawed TWG report would not help to bring the two sides together, but would only exacerbate the longstanding controversy between Dubna and Berkeley concerning the discovery and naming of elements 104 and 105.

However, all this was to no avail and the Phase 2 conclusions of the TWG, together with the already published^{13,7} Phase 1 report on criteria, were rapidly published in *Progress in Particle and Nuclear Physics*^{13,11} in 1992, with the title "Discovery of the Transfermium



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November 19, 1991

Aaldert H. Wapstra, Secretary
Transferrmium Working Group
NIKHEF POBox 41882
1009DB Amsterdam
The Netherlands

Via Fax (31) 20-5922165

Dear Aaldert:

We write with heavy heart and a keen sense of disappointment to comment on what we consider a seriously flawed report "Discovery of the Transferrmium Elements." On the basis of our discussions with you at the time of your visit to Berkeley we had expected a balanced and credible report. The report is so deficient in content that it is difficult to prepare a short comment, but we shall try.

As a prelude to our remarks we reiterate our vigorous protests, expressed during your Berkeley visit, against the arbitrary decision to include in your review all transferrmium elements--apparently because 100 is a unique number in our decimal system. Element 101 was discovered 36 years ago, based on exemplary experiments. You could just as well have discussed the dispute over the discovery of earlier elements such as 100, or even 93 (discovered 51 years ago), or perhaps 61 (discovered 46 years ago, see enclosed reprint).

In a bad start, the report downgrades the discovery of element 101 in 1955 by the Berkeley group. We have always regarded this research as a classic in that this very difficult discovery was made possible by **chemical** identification using the reliable ion exchange adsorption technique. Even the mass number and decay mechanism was designated correctly. Clearly, the poor half life determination was solely determined by the poor statistics from seventeen atoms produced from three consecutive experiments. The essence of the discovery was the chemical separation of element 101 from all other elements in 1955. The 1958 work was merely confirmatory, though valuable in its own right.

The report misses key points supporting the discovery of element 102 at Berkeley in 1958-59. For instance, it is now clear that ^{252}No was discovered in our 1959 work as a 3-second alpha emitter with a 33% S.F. branching decay. Fortunately, we had already prepared a draft account of the Berkeley work, prepared as a rebuttal to "The History of Element 102," prepared by Flerov et

Fig. 13.3. Complete text of Ref. 13.7. Fax sent to Aaldert H. Wapstra, Secretary of the Transferrmium Working Group, from Glenn T. Seaborg and Al Ghiorso, November 19, 1991.

al. for publication in Radiochimica Acta--we are therefore enclosing a copy of our "Comments on the Selective 'History of Element 102'."

The report's account of the discovery of element 104 is particularly disturbing to us and suggests the operation of a double standard in the evaluation of the work by the Berkeley and the Dubna groups. The Dubna identification of $^{260}104$, decaying by spontaneous fission, is obviously wrong so an attempt is made to credit the Dubna volatility work, marginal and inconclusive at best, which was performed after the Berkeley discovery experiments. On the other hand, no mention is made in the assessment section of the first meaningful chemical identification of element 104 at Berkeley in 1970 by the reliable ion exchange method. (Our recollection of the presentation in San Francisco in April 1968, which we attended, differs from the point of view you present.)

For element 106, no mention is made of the decisive clarifying experiments of Demin et al., which demonstrated that the spontaneous fission events were due in large part to an isotope of element 104. We regard this as evidence negating the Dubna claim for the discovery of element 106 by the observation of decay by spontaneous fission.

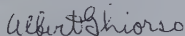
For element 108, we find it absurd that credence should be given to its discovery via the observation of remote decay descendants.

We find it curious that no mention is made (on page 4) of our publications among the "more elaborate expositions about the new elements." (We enclose a copy of a pertinent excerpt regarding element 102, as an example.)

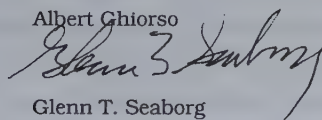
Had we been given the opportunity to comment on your report earlier we might easily have reached agreement on such modifications as we are as desirous as you to finding a solution to this problem. For example, we could agree to shared credit for the discovery of elements 102, 103, and 105.

We are making these suggestions rather hastily with the hope that these revisions can be made in your report before it is published. Otherwise, and reluctantly, we feel impelled to publish a more detailed rebuttal to your report when it appears.

Cordially yours,



Albert Ghiorso



Glenn T. Seaborg

Fig. 13.3 (continued)

Elements" under the authorship of the entire committee, R.C. Barber, N.N. Greenwood, A.Z. Hryniewicz, Y.P. Jeannin, M. Lefort, M. Sakai, I. Ulehla, A.H. Wapstra, and D.H. Wilkinson. The abstract stated in regard to Phase 1 that "...it was carried out in close consultation with the Laboratories." It further stated, in reference to the report on Phase 2, "It completes the work of the Group by applying the criteria of Part I on an element-by-element basis; it considers and analyzes all the pertinent literature and discusses in chronological and critical detail those papers considered important for the building up of confidence that each element had been put in evidence. This delineation of discovery profiles results, in some cases, in a sharing of the credit for discovery."

In early August 1992 while at GSI, Mike Nitschke took the initiative in proposing that a group of nine scientists, three from each of the three laboratories involved in the controversies over naming elements 101 through 109 (GSI, JINR, LBL/LLNL), get together to discuss how to achieve agreement. Mike proposed that if this "group of 9" could agree on a set of names, then it should be ratified at their respective laboratories and forwarded as a recommendation to the IUPAC/IUPAP. Shortly thereafter (August 10, 1992) Seaborg and Ghiorso received a fax from Paul Kienle, then Director of GSI, regarding naming in which he indicated that he, Nitschke, Y. Oganessian, and G. Münzenberg had arrived unanimously at a common proposal for the names of elements 101–109. However, the specific proposal was not included and Ghiorso and Seaborg indicated that based on the "meager information" in the fax they doubted that they could agree with it.^{13,12} They subsequently received a letter from Peter Armbruster written on August 10, 1992, in which he included the following list of names as the compromise proposal as of August 7: 102, joliotium, Jt; 103, lawrencium, Lw; 104, meitnerium, Mt; 105, kurchatovium, Ku; 106, rutherfordium, Rf; 107, nielsbohrium, Ns; 108, hassium, Hs; 109, hahnium, Ha. He stated that the Dubna merit for "cold fusion" is recognized by "nielsbohrium," and they could not go so far as to propose "Flerov," as discussed earlier. He further said that they were helping to solve the 104-naming problem by moving meitnerium

from 109 to 104 and moving hahnium to 109. Armbruster said that the list was based on the principle of trying to separate the naming of an element from its discovery, a principle which he realized Seaborg and Ghiorso had indicated they would not support. He further expressed his firm belief that the "time is overdue for a solution. The young generation has a right that the field does not suffer from our unsolved heritage. Otherwise, I am afraid we will have a serious problem to attract them to continue our work." He invited them to name three experts to meet with three each from Dubna and GSI in Paris to reach a solution — but the meeting would be of no use in case they could not agree with something like the proposed list! Seaborg and Ghiorso were also invited to a ceremony at GSI on September 7 to announce the names for elements 107–109, and if Dubna and Berkeley could agree on 102–106 these names could also be announced.

On August 11, 1992, Ghiorso and Seaborg replied^{13,13} to Armbruster expressing appreciation for his efforts, but disagreeing with the list of names in many respects and with much of the philosophy behind his efforts. They reiterated their belief that: (1) no attempt should be made to change the long-accepted names for elements 102 and 103 (nobelium and lawrencium), (2) it is not possible realistically to separate credit from discovery, and (3) names should not be suggested until discoveries have been confirmed. They further stated, "We would never agree to the naming of an element after Kurchatov (anymore than we would to the naming of an element after an American inventor of the hydrogen bomb) — and certainly not to the naming of element 105 after Kurchatov!" They did suggest that he consider naming one of his elements after a great Russian scientist such as Gamow, Goldanskii, or Landau.

They indicated that they could not meet him in Paris and strongly suggested that if he must make a naming announcement at GSI on September 7 he should revert to the original idea of using the "grand name 'meitnerium' for element 109" rather than duplicating the name "hahnium" by suggesting it for 109.

Subsequent to this exchange, many members of the 106 discovery group proposed names for element 106, since there was no controversy about its discovery (although it had not yet been confirmed). They also wished to have its naming “decoupled” from the rest of the naming process for the other elements.

Finally, on August 31, 1992, Ghiorso and Seaborg sent a short fax to Armbruster expressing appreciation to him and Mike Nitschke for trying to help find a compromise on the naming of elements 104 and 105, which failed primarily because of the Russian insistence on naming element 104 after Kurchatov.

Darleane Hoffman recalls^{13,14} the IUPAC meeting in Lisbon, Portugal, in August 1993, which she attended as an Associate Member and Past Chair of the Commission V.7 on Radiochemistry and Nuclear Techniques, and as a member of the US National Committee for the IUPAC. During that meeting, she contacted James Casey of the US, who was a member of the Commission 2.2 on Nomenclature of Inorganic Chemistry (CNIC) concerning whether and when heavy element naming and the TWG report would be considered by the CNIC. He referred her to the Chairman, Alan Sargeson, Australia, and she and Paul Karol (US, Commission V.7) contacted him. Sargeson said that Commission 2.2 would meet the following Sunday (August 8) at 2 p.m. Darleane met him at the door at that time and he said he doubted they would get to this matter at all. When she brought up the point about whether the report had been properly reviewed according to IUPAC regulations, he said they would investigate this. He also said his Commission did not have the required expertise to review it — they only considered nomenclature. In spite of this, the CNIC did not even discuss the matter with the officers and membership of Commission V.7, the only one in the IUPAC selected for specific expertise in nuclear and radiochemistry! Hoffman told Sargeson there was a letter from IUPAC President Alan Bard to Past President Jeannin (April 1993) expressing concern about publication in *Pure and Applied Chemistry* (PAC) without appropriate review although the Bureau had approved publication in *Progress in Particle and Nuclear Physics* by the IUPAP.

Normally, review by at least one internal reviewer and one external reviewer is required before publication in *PAC*, although Dr. M. Williams, Executive Secretary of the IUPAC, had indicated the report would be published in the August 1993 issue of *PAC*. He also stated that the TWG Report could now be considered by CNIC, which has the responsibility for recommending names! Hoffman urged Seaborg to contact Williams concerning appropriate review in order to try to resolve this issue before Alan Bard's term expired at the end of 1993 and the new president, Prof. K.I. Zamaraev (Russia), took office.

However, Parts II and III of the 1992 TWG "Discovery" article^{13.11} were published without such review in *PAC* in 1993,^{13.15} although responses were "invited" from the Berkeley, Dubna, and GSI laboratories. These responses were published (unedited) immediately following^{13.16} the TWG report. The response from Seaborg and Ghiorso^{13.17} was also published in *Progress in Particle and Nuclear Physics* in 1993, since no such opportunity had been afforded them by the IUPAP when the "Discovery" article^{13.11} was first published there.

13.4. Naming by the IUPAC and Protest

Thus the stage was set for another long period of dissent and confusion. Very quickly after the publication of the TWG deliberations, the CNIC of the IUPAC considered the naming of the transfermium elements at its meeting in Balatonfüred, Hungary, on August 31, 1994. Prior to their meeting, the CNIC had asked the three major groups concerned with the discoveries for their proposals and reasons for their choices of names for these elements. Responses were received from all three and were considered in the ensuing discussions. The CNIC decided unanimously to continue the practice of naming elements after appropriate scientists, places, and properties, but resolved specifically, "*an element should not be named after a living person.*" This specifically excluded naming element 106 "seaborgium," after Glenn T. Seaborg, as proposed by the undisputed discoverers, since Seaborg was obviously still alive and said, "I had no difficulty

proving it!" The CNIC also agreed to accept the conclusions of the TWG as one of the bases for selecting names as well as the suggestions from the three groups queried. All the names chosen came from these suggestions but "*not necessarily in the order suggested*," thus ignoring the historic right of discoverers to name the elements they discovered. These decisions meant not only throwing out "seaborgium," but scrambling the rest of the names in utter disregard of discoverers' suggestions or common usage by the heavy element nuclear chemistry community. For example, "rutherfordium," commonly used for 104, went to 106; "hahnium" went to 108, replacing "hassium," which although chosen by the undisputed GSI discoverers was dropped entirely. Furthermore, names were picked that had never been in common usage, such as "dubnium" for element 104 to replace "rutherfordium," and "joliotium" for element 105, replacing "hahnium." The confusion was compounded by shifting the Berkeley names of "rutherfordium" from element 104 to 106 and "hahnium" from 105 to 108! Thus, "hassium" and "seaborgium," both chosen by undisputed discoverers, were totally left out while the names "dubnium" and "joliotium," both proposed by Dubna, were adopted for 104 and 105, respectively. This showed a total disregard for the wishes of the discoverers of these elements as well as common usage in the heavy element nuclear chemistry community. This was probably not due to any ill-will on the part of the CNIC, but primarily a consequence of having no representatives from the nuclear chemistry heavy element community on the CNIC, nor were any, at least from the US or Germany, allowed to meet with the CNIC. The complete list of the selected names is shown in Table 13.2. The names for elements 101–103 approved by the IUPAC in 1971 were retained. These recommendations from the CNIC, chaired by Prof. Sargeson, were then accepted unanimously by the IUPAC Bureau at its September 17–18, 1994, meeting in Antwerp, Belgium, and approved for submission to the Interdivisional Committee on Nomenclature and Symbols as "definitive" for publication in *PAC*. The Bureau further decided to skip the five-month period required by the IUPAC by-laws to allow chemists around the world to

Table 13.2. Names Recommended by IUPAC/CNIC, August 1994. Approved by IUPAC Bureau, September 1994.

Element	Name	Symbol
101	Mendelevium	Md
102	Nobelium	No
102	Lawrencium	Lr
104	Dubnium	Db
105	Joliotium	Jl
106	Rutherfordium	Rf
107	Bohrium	Bh
108	Hahnium	Hn
109	Meitnerium	Mt

comment on nomenclature proposals and the names were published in *PAC*^{13.18} in 1994. This was justified in a report^{13.19} in *Chemistry International*, the IUPAC newsmagazine, on the basis of the "lack of agreement over some 30 years by the relevant laboratories." Again, this action was taken without any consultation with the US discoverers or those at GSI!

These "definitive" names resulted in a storm of protest and criticism from around the world, especially from the US. The US Organizing Committee for the IUPAC sent a letter protesting the failure of the IUPAC to follow their own rules for approval of nomenclature, and in November 1994 the Nomenclature Committee of the ACS approved a different set of names for use in their journals (see Table 13.3). A joint letter^{13.20} by former and current IUPAC members from the countries involved — Prof. Vitalii Goldanski (Russia), Prof. Jens V. Kratz (Germany), and Prof. Darleane Hoffman (US) — was sent to the IUPAC Directorate in March 1995. This letter, shown in Fig. 13.4, suggests principles that the IUPAC should establish before the naming process can be completed: (1) that the

Table 13.3. Names Approved by American Chemical Society, November 1994.

Element	Name	Symbol
104	Rutherfordium	Rf
105	Hahnium	Ha
106	Seaborgium	Sg
107	Nielsbohrium	Ns
108	Hassium	Hs
109	Meitnerium	Mt

historic right of the discoverers of a new element to name it be confirmed, and (2) that there should be no new, retroactive rule that elements cannot be named after living persons.

In response to this worldwide criticism, the new IUPAC President, Karol I. Zamaraev of Russia (President-elect H.A. Staab, who should have served as President, had resigned), asked the Executive Committee at its meeting in Reading, England, in April 1995 to reconsider the names adopted in September 1994 (Table 13.2). Those present were: President Zamaraev, Chair of the Executive Committee; Vice-President (President-elect) Prof. A.E. Fischli (Switzerland); Immediate Past President Prof. A.J. Bard (USA); Secretary-General Prof. G. den Boef (Netherlands); Treasurer Prof. J.M. Ward (UK); Elected Members Prof. R.D. Brown (Australia), Prof. J. Jortner (Israel), Dr. D.F. Eaton (USA). Also in attendance were Executive Secretary Dr. M. Williams (UK) and Deputy Executive Secretary Dr. J.H. Williams (UK).

Quoting from the official minutes^{13,21} of that meeting, "The President (Zamaraev) said that the recommendations approved by the Bureau had been duly endorsed by the IDCNS (Interdivisional Committee on Nomenclature and Symbols) and definitively published in *PAC*, as well as being released to national chemistry news journals. As a consequence of the heavy criticism arising, especially from the

March 29, 1995

To: Executive Committee of IUPAC:
Subject: Names and Symbols of Transfermium Elements

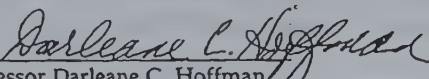
We have considered the names for the transfermium elements proposed by the Commission on Inorganic Nomenclature of IUPAC. We are very concerned that the historic rights of discoverers have not been honored.

We suggest that the following principles must be established at the highest level of IUPAC before the naming process can be completed.

1. IUPAC should confirm the historic right of the discoverers of a new element to name the element they discovered. If IUPAC refuses to approve it, the discoverers should be asked to propose a new name.
2. There should be no new retroactive rule that elements cannot be named after living persons. However, we recommend that the names be those of eminent scientists.

Signed:


Professor Vitalii Goldanski


Professor Darleane C. Hoffman



Professor Jens V. Kratz

Fig. 13.4. Copy of Ref. 13.20. Memo from Vitalii Goldanski, Darleane C. Hoffman, and Jens V. Kratz, to the Executive Committee of the IUPAC, regarding names and symbols of transfermium elements.

USA, Prof. Zamaraev had discussed the situation with the Vice-President and Secretary-General at the Secretariat (01/30/95). An historical overview and a proposal on how to proceed were then circulated to the Executive Committee (95.03.28) for consideration at the present meeting. After due deliberation the Executive Committee agreed to propose that the Bureau at Guildford should reconsider its decision at Antwerp, by deciding to adopt the recommendations as provisional, in full accordance with By-law 2.11. This would mean that the final ratification by the Council could not be made until it met in 1997.

"From his personal contacts in recent months with scientists from various countries, Prof. Zamaraev had the impression that a compromise on naming the transfermium elements could be found. Accordingly, he suggested that the Chairmen of the National Committees for the IUPAC in Germany, Russia, and the USA (or other responsible representatives of the NAOs from these countries) should meet at the Secretariat in late June 1995 for two days, together with the President of the IUPAC as a mediator, to discuss this delicate issue. The Chairman of the CNIC should also be invited to attend. Any agreed compromise should be submitted to this Commission for consideration at Guildford, with the request to advise the Bureau on how to proceed. If no compromise was reached, then, in due course, the IUPAC would continue to discuss the present recommendations of the Commission in accordance with statutory procedures.

"It was decided by the Executive Committee that the Executive Secretary should be the official IUPAC contact person to deal with enquiries about progress within the Union on names for the transfermium elements. However, information should only be released on the proposal for reconsideration by the Bureau at Guildford."

In an attempt to reach a compromise slate of names, this IUPAC *ad hoc* negotiating group, consisting of representatives of the National Adhering Organizations (NAO) of Germany, Russia, and the USA, duly met during the August 1995 IUPAC meeting in Guildford,

England. The USA representatives solicited information from Hoffman and Seaborg and others in the USA, but apparently those from Germany did not solicit information from the discovery group or their IUPAC nuclear chemistry members. The Russian NAOs apparently worked very closely with the Dubna group. For some reason, the *ad hoc* Committee went all the way back to element 101 and suggested the new "compromise" slate of names for elements 101 through 109 shown in Table 13.4. Indeed, they did reinstate "seaborgium" for element 106 — but essentially all the rest of the USA-proposed names were held hostage in return for retaining "seaborgium"! The rest of the proposal was, if anything, worse than the original set of names (see Table 13.2), primarily because none of the German or USA members were nuclear chemists. They seemed to feel that as long as each of the countries involved had a fair share of their proposed names chosen it did not matter for which element they were used.

Indeed, in an unprecedented step the Bureau did back down from its previous position and decided at the August 1995 meeting in Guildford that the 1994 recommendations of the CNIC which had

Table 13.4. Names Suggested by *Ad Hoc* IUPAC Committee of NAO Representatives, Guildford, England, August 1995.

Element	Name	Symbol
101	Mendelevium	Md
102	Flerovium	Fl
103	Lawrencium	Lr
104	Dubnium	Db
105	Joliotium	Jl
106	Seaborgium	Sg
107	Nielsbohrium	Ns
108	Hahnium	Ha
109	Meitnerium	Mt

been approved should revert to provisional status in compliance with the IUPAC by-laws, which stated that comments must be solicited over a period of at least five months. Comments from around the world were solicited and 19 national and regional centers were even designated to collect these comments until May 31, 1996. Prof. Zamaraev passed away in late 1995 and Prof. Fischli of Switzerland became President. In addition, Dr. Edwin Becker (USA) became Secretary-General and the IUPAC Secretariat was moved to the Research Triangle Park, North Carolina, USA. Dr. John W. Jost became Executive Director and Dr. Mo Williams retired as Executive Secretary. The term of Prof. A.M. Sargeson (Australia) as Chairman of the CNIC expired at the end of 1995 and Prof. G.J. Leigh (UK) became the new Chairman. All of these changes undoubtedly helped pave the way for a compromise solution to the heavy element naming problem.

During the comment period, there was an overwhelming swell of support for "seaborgium" for element 106, especially since there was no real precedent for not naming a chemical element after a living person. There was little support for naming element 104 "dubnium" rather than "rutherfordium." Many letters were sent out by Hoffman, Ghiorso, and Seaborg requesting support for the principles iterated in Fig. 13.4, namely confirmation of the historic right of discoverers to name the element they discovered and no new, retroactive rule against naming an element after a living person. Particularly important in this regard was the support of the Chinese and Japanese Chemical and Nuclear Societies.

13.4.1. IUPAC 1997 Approved the Names for Elements 101 Through 109

After receiving a multitude of comments, the CNIC in its August 1996 meeting proposed the "compromise" slate shown in Table 13.5. This list was subsequently approved at the IUPAC meeting on August 30, 1997, in Geneva, Switzerland. Rather curiously, in the article entitled "Names and Symbols of Transfermium Elements"

Table 13.5. CNIC/IUPAC Compromise Recommendation Approved August 30, 1997, Geneva, Switzerland.

Element	Name	Symbol
101	Mendelevium	Md
102	Nobelium	No
103	Lawrencium	Lr
104	Rutherfordium	Rf
105	Dubnium	Db
106	Seaborgium	Sg
107	Bohrium	Bh
108	Hassium	Hs
109	Meitnerium	Mt

(IUPAC Recommendations), published in 1997,^{13,22} the CNIC recommended that element 105 be named “dubnium” in honor of the Dubna laboratory for playing “a key role in developing the experimental strategies used in synthesizing several transfermium elements.” In addition, the TWG assigned shared credit (Table 13.1) for the discovery of element 107 to the GSI and Dubna teams. Because of this, the CNIC recommended the name “bohrium” for element 107, in recognition of the previous proposal of “nielsbohrium” for element 105 by the Dubna group.

We can generally accept these approved names, but recommend the continued use of “hahnium” for element 105 due to its extensive use in the literature over the last 25 years. The American Chemical Society’s Committee on Nomenclature endorsed this list, but also with reservations about the name “dubnium” for element 105 because of the long use of the name “hahnium” for this element: “In the interest of international harmony, the Committee reluctantly accepted the name ‘dubnium’ for element 105 in place of ‘hahnium,’ which has had long-standing use in literature. We are pleased to note that

'seaborgium' is now the internationally approved name for element 106."^{13.23}

But we still believe the credit for the discovery of the transfermium elements was erroneously assigned by the TWG and should be as follows: element 101, Berkeley (1955 group)^{13.24}; element 102, Dubna^{13.25}/Berkeley^{13.26}; element 103, Berkeley^{13.27}/Dubna^{13.28}; elements 104–106, Berkeley^{13.29–13.31}; elements 107–109, GSI.^{13.32–13.34}

13.5. Names for Elements 110, 111, and 112

For indexing purposes, the CNIC in 1979^{13.35} recommended the use of three-letter "systematic names" based on 0=nil, 1=un, 2=bi, 3=tri, 4=quad, 5=pent, 6=hex, 7=sept, 8=oct, and 9=enn for elements of atomic numbers greater than 100, until the approval by the IUPAC of so-called trivial names. Thus, technically speaking, according to the IUPAC, the names for 110, 111, and 112 will be "ununnilium" (Uun), "unununium" (Uuu), and "ununbium" (Uub) until names for them are suggested and approved for these. Perhaps with a view to avoiding such an eventuality, as early as September 1996, Prof. G.J. Leigh, the new Chairman of the CNIC (who had also been instrumental in negotiating the 1997 solution for naming the transfermium elements shown in Table 13.5), sent out a confidential letter^{13.36} asking the laboratories at GSI, Dubna, and Berkeley whether they regarded the synthesis of elements 110 and 111 as unequivocally established, to whom priority for synthesis should be assigned, and whether the discoverers should be asked to suggest a name. Seaborg received this letter for LBNL and replied. After the announcement by GSI of the discovery of element 112, Leigh sent another letter (October 13, 1997), to which Seaborg replied for LBNL^{13.37} (Fig. 13.5), suggesting that the GSI group should propose names for elements 111 and 112 and that the GSI, Dubna/Livermore, and Berkeley groups each suggest a name for element 110 from which the Commission on Nomenclature of Inorganic Chemistry could choose. He further indicated that the Berkeley group supports the choice of



OFFICE OF THE ASSOCIATE DIRECTOR-AT-LARGE
GLENN T. SEABORG

November 3, 1997

Dr. G. J. Leigh
The School of Chemistry, Physics
and Environmental Science
University of Sussex
Falmer, Brighton BN1 9QJ
ENGLAND

Dear Dr. Leigh:

This is in reply to your letter of October 13, 1997.

I agree that, as the discoverer of elements 111 and 112, the GSI group should suggest the names.

In the case of element 110, I think that your second alternative is the only feasible approach, namely that the GSI, Dubna, and Berkeley/Livermore group each suggest a name from which the Commission on Nomenclature of Inorganic Chemistry can make a choice.

Our Berkeley group believes that element 110 should receive the name "Hahnium," in honor of the scientific giant "Otto Hahn." This also has the advantage that it would then not be necessary for so many of us to continue to use the name "Hahnium" for element 105 to honor Otto Hahn.

Sincerely yours,

Glenn T. Seaborg

GTS/njt

c: A. Ghiorso
'D. Hoffman'

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Fig. 13.5. Copy of Ref. 13.37. Letter from Dr. Glenn T. Seaborg to Dr. G.J. Leigh, November 3, 1997.

"hahnium" for element 105 in honor of Otto Hahn, codiscoverer of nuclear fission.

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Chapter 14

Searches for the Superheavy Elements

14.1. Introduction

Although the existence of superheavy elements (SHE) had been predicted by John Wheeler^{14.1} as early as 1955 and discussed by Gertrude Scharff-Goldhaber^{14.2} in 1957, the big wave of interest in SHE was not triggered until nearly ten years later, by predictions by Myers and Swiatecki^{14.3} and H. Meldner^{14.4} of an Island of Superheavy Elements well beyond uranium. Subsequently, theoretical studies based on newly developing theories of nuclear structure and shell corrections to the liquid drop model by Strutinsky,^{14.5} Nilsson,^{14.6} Fiset and Nix,^{14.7} and others indicated an island of nuclear stability around element 114. A fanciful depiction of the search for SHE is shown in Fig. 14.1. This region was initially believed to be stabilized by spherical shells at 114 protons and 184 neutrons, which were nearly as strong as those at 82 protons and 126 neutrons found in the stable lead region — to the extent that a “Magic Island” or “Island of Stability” with half-lives as long as a billion years might exist! (Toroidal shapes were even postulated^{14.8} to be possible above $Z=134$.) These predictions raised the possibility that the most stable part of the Island might have been formed in the last nucleosynthesis in our solar system nearly five billion years ago and that the very long-lived elements might still exist in nature. This idea prompted many researchers to look for SHE in natural ores as well as to try to produce them at accelerators.

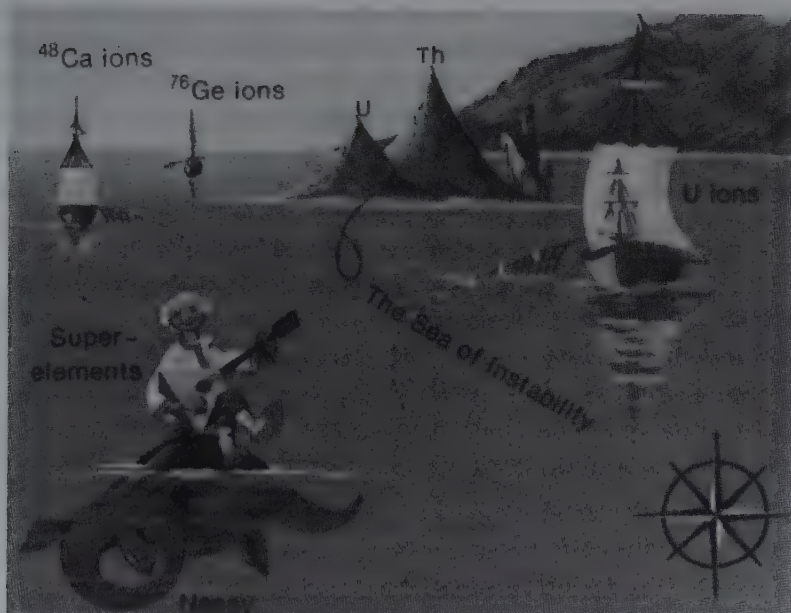


Fig. 14.1. A fanciful depiction of the search for SHE, from G.N. Flerov, 1976.

There have been a number of reports on the discovery of SHE both in nature and by production at accelerators. These announcements have always been exciting, at least for a short time, until either it became obvious that they were in error or until confirmatory experiments failed to confirm the original reports. It is not surprising that many "false discoveries" should be made in this area, because the discovery of a far distant whole new region of the periodic table of the elements requires a long extrapolation from known chemical as well as nuclear properties. The discovery of such a new region of the periodic table would, indeed, be a major scientific advance that would be widely acclaimed by the lay public as well as by fellow scientists.

Within a few years many experiments had been performed to search for the hypothesized new group of elements. Very early

experiments were conducted at accelerators and it became apparent that if the Island existed at all it was not going to be easy to reach. But there was always the possibility that the most stable part of the Island might have already been formed in nature and that the SHE might still exist. Many searches in nature were started with this in mind. Moreover, even if the SHE had decayed, it might still be possible to detect products from their SF decay, such as the stable isotopes of the rare gases krypton and xenon.

14.2. Early Searches for SHE in Nature

Günter Herrmann published a review^{14.9} of these early searches for SHE in nature as of mid-1974, commenting that although many extremely sensitive detection techniques had been developed, no positive results had yet been obtained. Among the earliest searches were those conducted at Berkeley between 1968 and 1972 by Stan Thompson's group.^{14.10} In the first searches they looked specifically for eka-platinum, element 110, using low background counting techniques and sensitive analytical methods. Their results were negative, corresponding to a concentration of $< 10^{-11}$ g/g. Later, Eli Cheifitz, Richard Jared, E.R. Giusti, and Thompson decided to look for the high multiplicity of neutrons expected from the SF of spherical SHE isotopes in the region of element 114. They used their high efficiency, large liquid scintillator system in an attempt to detect SF events in which unusually large numbers of neutrons were emitted. In order to reduce effects from the cosmic ray background, they located the detector some 250 m deep in a cross passage inside the Bay Area Rapid Transit (BART) system tunnel leading from Berkeley to Orinda. (Glenn Seaborg recalls that in May 1970 he and Helen, together with Stan Thompson and Eli Cheifitz, walked some 7000 feet into the BART tunnel to the site of the neutron coincidence counting apparatus situated in the cross tunnel.) Elements 111 through 114 were predicted to behave as eka-gold, -mercury, -thallium, and -lead. Based on these predictions of chemical properties, they surveyed more than 40 large samples of ores, natural minerals such

as galena and gold nuggets, manganese nodules from the ocean floor, moon rocks, and placer platinum, but found no evidence for increased neutron emission. A photo of these researchers with the gold nuggets which were surveyed in the neutron detector system is shown in Fig. 14.2. Assuming a half-life of 10^9 years, they set a limit of $< 10^{-14}$ moles of SHE per mole of sample.



Fig. 14.2. Left to right: Eli Cheifetz, Richard Jared, Stan Thompson, and E.R. Giusti with gold nuggets.

In 1969, Flerov and coworkers reported^{14.11} detecting fission tracks in lead glass which they attributed to the possible decay of SHE. Based on additional observations of fission events in lead ore samples, they concluded they had found SF events with an apparent half-life of 4×10^{20} years, which they attributed to SHE. However, P.B. Price, R.L. Fleischer, and R.T. Woods^{14.12} could not observe any fission tracks in old lead- and gold-rich minerals and set limits which were in contradiction with the Flerov results.

After Darleane Hoffman and coworkers^{2,11} reported the separation and detection of minute quantities of ²⁴⁴Pu in bastnasite ore in 1971, Darleane remembers often being asked why she didn't try to chemically separate SHE from natural sources. She says, "I replied that finding plutonium was difficult enough even though I already knew much about its chemistry and where it might be concentrated, but trying to look for an exceedingly long-lived element whose atomic number and chemistry I could only guess at seemed nearly impossible!"

14.3. Early Searches for SHE at Accelerators

The first attempts to produce SHE "artificially" were conducted in 1968 by Stan Thompson and coworkers^{14,13} and Al Ghiorso and coworkers^{14,14} using reactions of heavy ion projectiles with heavy actinide targets, e.g. $^{40}\text{Ar} + ^{248}\text{Cm} \rightarrow ^{288}114^*$. No SHE were detected and only limits could be set on the production cross sections and half-lives. In January 1971, A. Marinov *et al.*^{14,15a} published an article in *Nature* claiming production of element 112, eka-mercury, after they observed SF events in a mercury fraction chemically separated from the products of a long irradiation of tungsten with 24-GeV protons. The production mechanism was presumed to be secondary reactions with suitable targets of the heavy recoil products from the interactions of the high energy protons. However, later experiments showed^{14,15b} that ~70% of the SF activity was due to contamination from ²⁵²Cf and after its subtraction the residual fission kinetic energy spectra seemed to differ considerably from those of known actinides. However, subsequent attempts^{14,16} by members of that group to repeat the initial results were unsuccessful. Also in 1971, Bimbot *et al.*^{14,17} in Orsay, France, bombarded ²³²Th with 500 MeV ⁸⁴Kr ions (the compound nucleus would be [³¹⁶126]*), and reported finding very high energy (13–15 MeV) α -particles among the reaction products. The cross section would correspond to about 10 mb, with the half-lives being between about a ms and a minute. They suggested these might come from isotopes of SHE. They also said their results

were an additional indication for the existence of the SHE reported earlier by Marinov *et al.* However, subsequent, more sensitive experiments proved to be negative.

In 1972, Flerov and Oganesian^{14.18} reported detecting an ~ 150 -d SF activity in sulfide fractions containing osmium and bismuth separated from the products of long bombardments of ^{238}U with a variety of ions as heavy as ^{136}Xe . However, the average number of neutrons per fission was typical of actinides rather than SHE, so, once again, another report seemed unlikely. However, attempts were initiated to try to produce SHE in uranium-uranium collisions at the UNILAC in Darmstadt, Germany, and in reactions of ^{48}Ca projectiles with ^{248}Cm at the SuperHILAC at Berkeley.

14.4. Summary of Results up to 1978

Some years later, because of the increasingly intense interest in SHE around the world, a large International Symposium on Superheavy Elements was organized in 1978 in Lubbock, Texas, USA. There was so much interest in the subject that most of the principal scientists working in the field attended, as did a large audience of other interested scientists. Some 15 countries and 50 different institutions were represented. Both Darleane Hoffman and Al Ghiorso remember attending. In fact, Al chaired the first session, on "Searches for SHE at Accelerators," and Darleane presented a paper (Ref. 14.19, p. 89) on "Neutron Multiplicity Measurements for SF of Cf and Fm Isotopes and Relevance to Neutron Emission for SHE."

John Wheeler, then Director of the Center for Theoretical Physics at the University of Texas in Austin, was unable to attend, but sent an exceedingly interesting letter to the Symposium (Ref. 14.1, p. xix), which is reproduced here:

"Warm greetings to colleagues and participants in this important conference timed in happy (anticipation of the?) 40-year celebration of the historic discovery of Hahn and Strassmann. We celebrate also the unique insights that fission and the search for superheavy

elements continue to give us into the structure of the atomic nucleus. Nowhere more strikingly than here do we see the coupling of individual particle and collective modes of motion. Nowhere more than here does the nucleus continue to surprise us with new physical effects and reward us with new understanding. As we continue our research on superheavy elements, with all it has to tell us about fission, we cannot forget the responsibilities and opportunities that go with this work. No technical development has had more to do with keeping the peace for the past third of a century than the world's fears of fission devices and fission-initiated fusion devices. Today fission provides the electric power for tens of millions of people throughout the world, and tomorrow it will be hundreds of millions. Nowhere more than in our branch of science, truly international from the very beginning, are the opportunities greater for us to work for that 'open world' that Niels Bohr told us is the only true guarantee of peace among nations."

An opening history and perspective of the search for SHE was given at the Lubbock Symposium on Superheavy Elements (Ref. 14.19, p. 10) by Dr. O.L. (Lew) Keller, Jr. [Director of the Transuranium Laboratory at the Oak Ridge National Laboratory and a member of the IUPAC Ad Hoc Committee appointed in 1974 (see Chapter 13) to try to resolve the element 104, 105 naming and discovery controversies]. He reviewed some of the predictions of chemical properties of the SHE and their expected ground state electronic configurations as well as oxidation states, ionic radii, and other properties. For example, element 114 was expected to be eka-lead, but to be somewhat more volatile. He gave an excellent overview not only of the history, but of the status of the current nuclear theory and current searches for SHE, both in nature and at accelerators. Searches for SHE had already been made using the accelerators at Berkeley, Darmstadt, Dubna, and Orsay. At Berkeley,^{14.20, 14.21} investigations had been undertaken of the reaction of ^{48}Ca projectiles with ^{248}Cm targets, believed to be the reaction of choice for making

SHE because it has the highest neutron-to-proton ratio available. But the experiments were all negative, netting only a limit on the cross section of 0.1 nb (10^{-34} cm²) for half-lives between about 0.1 d and 200 d. At the UNILAC at Darmstadt, Herrmann and colleagues were using their unique uranium beams to bombard uranium targets, but also to no avail.

Although the searches in nature had not yielded positive identifications of SHE, investigations of anomalous stable xenon ratios in the Allende meteorite by Anders and coworkers^{14,22} led them to postulate that they might be due to SF of an extinct SHE. Although this conclusion was model dependent and rather controversial, it did stimulate a number of experimenters to look more closely at Allende as well as other carbonaceous chondrites. Bob Gentry^{14,23} early began his investigation of giant halos in mica and was engaged in trying to come up with an acceptable explanation for their origin.

Keller pointed out in his Overview at the Lubbock meeting that even if SHE had not yet been discovered, their impact on the field of nuclear science had been great, furnishing a large part of the impetus for the construction of the new heavy ion accelerators at Darmstadt, Berkeley, and Oak Ridge, and helping to furnish the motivation for the development of theories of high energy interactions of heavy nuclei. He closed his Overview with the following remarks, which seem worth repeating here, lest we become too critical of some of the initial reports of discoveries of SHE which were eventually discredited or remain unsubstantiated: "We are still groping in a largely unexplored and poorly understood intellectual frontier. Right now this frontier is called superheavy elements; but, of course, it may turn into something else just as exciting. These transformations have occurred in the past, as when Fermi's research into transuranium elements turned into the discovery of fission through the inspired and meticulous chemistry of Otto Hahn." (And Fritz Strassmann.) It is well for us to remember that even the best of scientists make mistakes, for example Fermi's erroneous report of the discovery of the first transuranium element^{1,1} and his group's

nondiscovery of fission, probably because they had no “chemist” in the group!

Detection of neutrons was often used in these searches, and although it is a very sensitive detection method, it suffers from being nonspecific, as we discussed in Chapter 9. This is especially troublesome in the search for very small concentrations of SHE, because by the time the searches for exceedingly small numbers of neutrons from the SF of SHE were undertaken, spontaneously fissioning actinides were being used as target materials at many accelerators in the US, Russia, and elsewhere, and had also been deposited as fallout from atmospheric nuclear weapons tests and incorporated in minute quantities in a host of environmental samples. Consequently, in any neutron signals detected, the possible contribution from contaminants had to be considered. Groups at Dubna, Berkeley, Los Alamos, Livermore, and Oak Ridge all had neutron multiplicity counters which were used in searches for the SHE, with identification being based on the detection of the neutrons from SF of the SHE with the high multiplicity of around 10 neutrons per fission on the average, $\bar{\nu}_f$, predicted by Nix^{14.24} in 1969, compared to $\bar{\nu}_f$ of about 2 for uranium and 3.7 for ²⁵²Cf.

Flerov and his group^{14.25} at Dubna had mounted one of the most comprehensive searches for SHE in nature. They used a neutron multiplicity counter placed deep in a mine to reduce the background in order to look for neutrons from SHE in nature. They reported finding neutrons above background in samples of the Allende meteorite and in samples which Ivo Zvara had processed from 100 M³ of Cheleken brine water. He had concentrated the sample on 170 kg of anion resin, and after further processing, prepared a final acid sulfate precipitate in which about 1 SF per day was detected. However, the average multiplicity was lower than that of 10 predicted by Nix. In her paper (Ref. 14.18, p. 89) at the Lubbock Symposium on SHE, Darleane compared the neutron multiplicity data taken for two fractions from this processing with those for the ²³⁸U and ²⁵²Cf standards measured in their system and with hypothetical multiplicity distributions calculated for their system for $\bar{\nu}_f$'s of 5 and 10. She

found that most of the data for their samples were consistent with known actinides or possibly with $\bar{\nu}_t = 5$, but that SF with $\bar{\nu}_t = 10$ was excluded. However, she also pointed out that based on experimental measurements^{14,26} at Los Alamos the average number of neutrons, $\bar{\nu}_t$, emitted from high total kinetic energy, symmetric mass splits in the SF of ^{257}Fm is only 1.1. That is, only a few neutrons (1–2) per fission are emitted in the symmetric fission of ^{257}Fm in which two near-spherical fission products are produced. She cautioned that these data suggest that in the SF of a SHE into near-spherical fission fragments the fragments may not emit the very large number of neutrons that has been predicted theoretically by Nix and others for SHE. She calculated that $\bar{\nu}_t$ may be only about 5 for SF of $^{298}114$ into two symmetric fragments.

George A. Cowan, Division Leader of the Chemistry–Nuclear Chemistry Division at Los Alamos, gave the Symposium Summary. George was the person primarily responsible for urging Darleane Hoffman to undertake the search for ^{244}Pu in nature when he was group leader of the Radiochemistry Group at Los Alamos. He had also been interested for many years in trying to recover heavy elements produced in underground nuclear tests, and as all of those at Los Alamos and Livermore who engaged in these searches were well aware, this was an extremely difficult task. As they tried to find elements heavier than fermium which might be present in extremely small concentrations in kilograms of Nevada “dirt,” they became cognizant of the care that must be exercised to make sure there is no contamination from other sources, one of the more troublesome of these being from the α -emitters from the ubiquitous thorium and uranium natural decay chains. Darleane remembers the “cardboard box” incident when she kept seeing the decay of rather short-lived alphas at the level of a few counts per hour. She observed, after repeated counting of the samples, that the activities would “reappear” when the samples were reinserted in the Frisch grid chambers after having been stored for sometime in their cardboard boxes! At first she thought these activities might be due to impurities in the counting gas mixture, but after some more detective work it appeared that

the activities “emanated” from natural thorium/uranium present in the cardboard and that these activities deposited on the platinum sample disks!

In his closing Summary of the Symposium, George Cowan said, *“We may, indeed, be approaching a resolution of these mysteries, but this is not the meeting which I hope to attend someday when the rapporteur can announce an historic breakthrough in our level of understanding.”* The searches at accelerators had provided largely negative news and the plans for the future seemed to lie in pushing the limits to shorter half-lives and lower cross sections. Herrmann^{14.9} predicted that the increases in flux and fluence levels should permit measurements of cross sections for the U + U reaction down to about 10^{-35} cm² compared to their recent cross section estimates for the formation of element 114, which were up to ten times larger than that. The experimental situation for searches in nature seemed to be that there were groups of α -particles of both anomalously high and low energy, anomalously long fission tracks possibly from SF in radiogenic lead, and high neutron multiplicity fission events in hot springs extracts and chondritic meteorites. Cowan observed that although careful analyses of the experiments permit alternative explanations for some of the observations, not all of the data can be easily discounted. Thus he pointed out that the present observations could be used as screening techniques to identify interesting bodies of terrestrial material large enough to permit more detailed investigations and the chemical separation of the characteristic groups and the eventual separation of larger quantities of the SHE. He suggested that the bombardment of ²⁴⁸Cm with ²³⁸U should be carried out and that if that proved unsuccessful, attempts should be made to recover much larger quantities of the ²⁵⁰Cm produced in underground nuclear tests, another difficult task which has never been undertaken.

14.5. Searches for SHE Since 1978

Subsequent to the Symposium, a large collaboration of scientists, including Armbruster’s group and Gaggeler’s group from GSI,

Herrmann's group from the University of Mainz, and H. Dornhöfer from the University of Göttingen in Germany; Ghiorso's and Seaborg's groups from the Lawrence Berkeley Laboratory and Hoffman, Mac Fowler, and Bill Daniels from Los Alamos National Laboratory in the US, and Hans von Gunten from the University of Bern, Switzerland, was formed to thoroughly investigate the fusion of ^{48}Ca projectiles with ^{248}Cm targets leading to the compound nucleus $^{296}116^*$. Projectile energies close to the reaction barrier were used in order to try to keep the excitation energy as low as possible so as to minimize losses due to prompt fission. This was considered to be one of the most promising approaches to the production of SHE, but previous attempts at Berkeley (Ref. 14.19, p. 34; Refs. 14.27, 14.28) and at Dubna^{14.29} at bombarding energies corresponding to excitation energies of the compound system of 33–53 MeV had all been negative. An extra energy or "extra push" of about 20 MeV over the calculated fusion barrier was thought to be necessary in order to overcome entrance channel limitations on the fusion process, but more recent work of Armbruster had indicated that the dynamic hindrance for this system should be small. So this new search was initiated at energies much closer to the Coulomb barrier, calculated to be about 235 MeV. Experiments were performed at the SuperHILAC at Berkeley and at the UNILAC at GSI, Darmstadt. Both the recoil fragment separators, SASSY at LBL and SHIP at GSI, were used to search for short-lived nuclides. In addition, a variety of different radiochemical techniques were employed to search for longer half-lives. (See Fig. 14.3.) In the radiochemical searches, standard off-line chemical techniques were used for longer-lived species and fast on-line separations of the order of seconds were used for short-lived species. Three different separation approaches were used, based on the predicted chemical properties: (1) elements 112 through 116 were thought to be volatile at temperatures up to 1000°C (Pb-like); (2) element 112 or 114 might be gaseous at room temperature (Rn-like); (3) elements 108 through 116 were expected to form strong anionic bromide complexes in aqueous solution (Pt-like). The on-line systems OLGA, ARCA, and a cryo-

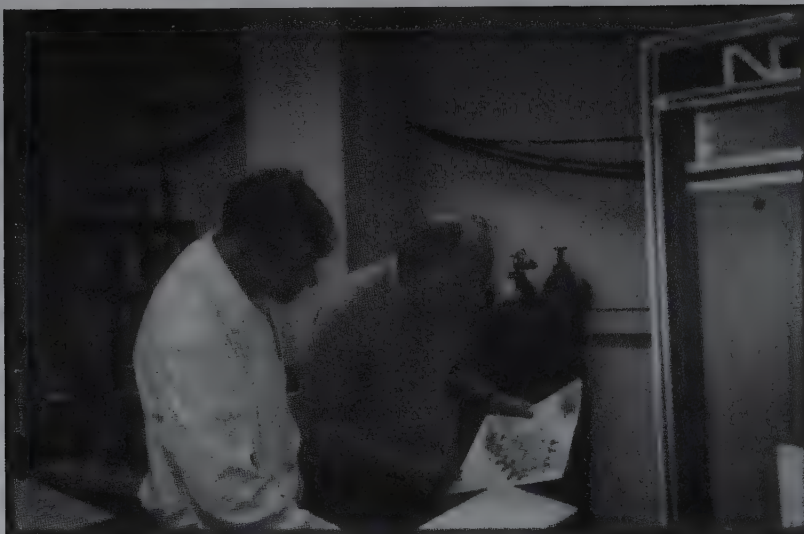


Fig. 14.3. Profs. Hans von Gunten (University of Bern, Switzerland) and Günter Herrmann (University of Mainz, Germany) comparing the puzzling results with the chart of the nuclides.

genic system to condense Rn-like noble gases were used as appropriate. Utilizing all these techniques, a range of SHE half-lives of 14 orders of magnitude — from 1 ms to 10 years — was covered! Although this broad range of half-lives was covered and excitation energies of 16–40 MeV were examined, the collaboration^{14,30} ultimately reported that there was no evidence for production of SHE with cross sections larger than 10^{-34} – 10^{-35} cm² (100–10 pb).

Searches for naturally occurring SHE in Atlantis II hot brine were reported in 1979 by Flerov *et al.*^{14,31} and in 1981 by Halperin and coworkers,^{14,32} but with negative results. Members of Reinhard Brandt's group from Phillips University, Marburg, Germany, working at the Los Alamos National Laboratory during several summers, together with members of Darleane Hoffman's team in the LANL Radiochemistry Group, also searched for known long-lived actinide

isotopes and set very low limits^{14.33} for their occurrence in these brines as well.

14.6. Some Notable “Nondiscoveries” of SHE

14.6.1. Giant Halos and the Evidence for the “Discovery” of Elements 116, 124, 126, and 127

One of the most renowned cases of the “discovery” of SHE in nature came about with the announcement in 1976 by an Oak Ridge–University of California, Davis–Florida State group^{14.34} of evidence for the presence of element 126 and possibly for elements 116, 124, and 127 as well in several “giant halos” in mica samples. Robert Gentry, a staff member of Columbia Union College, who had been a visiting scientist in the Chemistry Division at the Oak Ridge National Laboratory since 1969, had long been interested in the origin of these halos. He actually met with Glenn Seaborg, then Chairman of the AEC, in December 1968 to show him pictures of pleochroic halos, presumably caused by a radioactive species. These halos usually appear as concentric rings around radioactive inclusions contained in various minerals, such as mica. Commonly, they are found surrounding microscopic inclusions of monazite containing uranium and thorium and their natural decay chains and are referred to as U/Th halos. Some of these had diameters, which, if attributed to a radioactive origin, would correspond to α -energies up to 14 MeV, much higher than any known radioactivity. Gentry’s hypothesis was that this might be an indication of the present or past existence of SHE in these inclusions. According to a paper of Lew Keller, Seaborg then called their attention at Oak Ridge to Gentry’s work in collecting giant halos and studying their origin.

The radiation damage from these α -particles in the course of millions of years can be observed under the microscope by cutting and polishing surfaces to show a series of circles whose diameters correspond to the ranges of the members of the radioactive series. In addition to these halos it had long been known that there existed another class, the so-called “giant halos,” and Gentry suggested that,

since it would require α -energies of 12–14 MeV to produce them, these might be produced by very long-lived superheavy element α -emitters. The question was: Could one identify these atoms by bombarding a minuscule single giant halo inclusion with low energy protons?

A team headed by Tom Cahill at the University of California at Davis had developed a very sensitive proton-induced X-ray technique to identify each element by its characteristic L X-rays (the so-called PIXIE method) using the rebuilt 60-Inch Cyclotron (transplanted from Berkeley). In this case they found that they could not use the K X-rays of the superheavy elements, because their predicted energies would be masked by numerous γ -ray transitions from the rare earth elements which were plentiful in the monazite samples. On the other hand, SHE L X-ray transitions with energies in the range of the order of 25 keV are in a window — between the most energetic L transitions of uranium (≈ 21.3 keV) and the least energetic K transitions of the rare earths (≈ 33 keV) — where the background is orders of magnitude smaller. In addition to this advantage they gained a factor of 10^3 in cross section to produce the L vacancies by proton bombardment. The energy of the proton beam was constrained to be within the limits of 4.5 and 5.8 MeV, the former value to insure that they could excite X-rays deep in an inclusion and the latter to avoid the background from a $^{27}\text{Al}(\text{p},\text{n})$ reaction. One of their big problems was to bombard only a particular tiny inclusion and not the area surrounding it, so the project was soon moved to the Florida State University Tandem Van de Graaff, where they had developed a very highly focussed proton beam that could be used as a microprobe. It was not trivial to keep the beam on target when that target was only 50–250 mm in diameter, but they managed most of the time.

The finely focussed proton beam was directed at a given halo and the X-ray spectrum measured with a nearby LN-cooled Si crystal. After a lot of runs with a number of halos they decided that they were seeing the characteristic L X-rays from element 126 and probably other superheavy elements, even though their data were somewhat

erratic, not very reproducible, and always obtained in the presence of a very high background. They had to use large proton currents and since the beam was concentrated into a tiny area, damage to the halos became a problem. The experimenters assumed that this was the reason for their nonreproducible results. After presumably careful analysis, however, they announced the discovery of the SuperHeavy Elements. If they were right, they had found something that was some 20 elements higher in atomic number than the heaviest known element at that time! This was momentous news to which all scientists would pay attention!

Al Ghiorso doesn't remember who phoned him with the news of this absolutely fantastic "discovery" of SHE in nature, but he remembers it as being electrifying only for a short time. The giant halos that had been analyzed came from monazite sands — and he thought that this meant that there would be at least kilograms of SHE available! Obviously, it would be a great target material for producing other superheavy elements, both higher and lower in atomic number, and would open up a vast new region of the elements for exploration. The Berkeley scientists in the field were buzzing with excitement and speculation.

After some thought Al decided that there must be something wrong with the experiments. He and others felt that they could not be right for a number of reasons. However, in a rather short time they were surprised to hear that there was an announcement from the University at Davis of an official reception and a lecture on the subject by the "Discoverer of the Superheavy Elements, Tom Cahill." This lecture was attended by Seaborg and in the question period after the lecture it was obvious that no other person was prepared with challenging or penetrating questions, so it soon turned into a debate between him and Cahill. Seaborg describes the episode in this way in his *Journal* entry of July 12, 1976:

"I left my LBL office at 2:30 p.m. and drove to Davis to attend the Crocker Nuclear Laboratory Research Progress meeting in 66 Roessler Hall, at which Prof. Thomas A. Cahill was scheduled to speak on "Evidence for Element 126 (Bicentennium?) and Some

Other Superheavy Elements.” Cahill described the experiments of his group rather thoroughly, making a special reference to the fact that people have mistakenly looked in nature for element 114 rather than 126, which should have been a more obvious place to look. The room was packed, with many people sitting in the aisles and standing in the back.

“John Jungerman presided, and when he asked for questions at the end, I came to the front of the room and made some comments and asked some questions. I complimented Cahill on the work, said that he had given an extremely exciting report, declaring that right or wrong, this work has given a tremendous boost to the field of superheavy elements. I said that he and his assistants had certainly proved the existence of 27.25-keV radiation, but the question of whether it might be due to prompt or delayed gamma ray emission must be settled beyond doubt. Cahill said that it is not due to delayed gamma ray emission as shown by its nonobservance when the beam is off, and they have tested many elements for prompt gamma ray emission with negative results. I asked how he explains the 14-MeV α -particles, other than their coming from an extinct element 164 with many intervening alpha decays; he had no explanation and said this is under study. I pointed out that the concentration of 126 to an extent of 1 part in 10^4 indicates large quantities which should have been found as a result of meticulous spectrographic analyses, which have been traditional in chemical work in the past. He replied that the quantities are not so large because the 126 occurs only in rare inclusions identified by the giant halos and may occur only in a couple of ore deposits in the entire world. I asked how he explains the separation of elements 124 and 126 in the inclusions; Cahill pointed out that even the concentration of uranium varied upon scanning the surface of a crystal or inclusion.

“I added the comment that 124 and 126 might differ in their higher oxidation states and hence be readily separated. I asked whether he is worried about having four magic numbers — $Z=126$, $N=228$, $Z=114$, $N=184$ — and Cahill replied that he was, and feels that perhaps 116 is not really present. I pointed out that his problem

is exacerbated by the need to have β -stable nuclides and that this is difficult for element 116 and even for element 124 (which is neutron-excessive with 228 neutrons) and he conceded that this is a problem, but perhaps a not-too-difficult one."

It turned out finally that the pseudo-X-rays that were seen were actually mostly due to a 27.3-keV photon from ^{140}Pr produced via the (p,n) nuclear reaction induced by the interaction of the 5.7-MeV protons with ^{140}Ce in the inclusions being bombarded. This was investigated by the Florida State part of the team, who began a systematic effort in July 1976 to find all possible nuclear gamma rays that might be produced by 4.7–5.7-MeV protons, but had not yet been cataloged. J.D. Fox and coworkers reported^{14,35} finding one gamma ray that could interfere with the claimed 27.3-keV X-ray from element 126. However, they proposed that the ^{140}Pr photon was not strong enough to alter their earlier conclusions, unless the cerium was concentrated in a thin layer near the surface of the monazite inclusions.

Darleane remembers, "I had previously been involved in studies of short-lived cerium and praseodymium isotopes at Los Alamos and was immediately rather skeptical about the reported SHE with an X-ray energy of 27.2 keV, about the same energy as the low-lying levels in praseodymium isotopes. But we were all very excited about it and held a meeting to discuss what nondestructive testing could be conducted to determine the Z and perhaps even the mass and how more giant halos might be obtained. However, it seemed too good to be true that in addition there was some evidence for elements 116, 124, and 127!" Element 126 would be a "superactinide" and might be chemically expected to concentrate in monazite which is $(\text{Ce, La, Th})\text{PO}_4$ and can cocrystallize other lanthanides and actinides.

14.6.2. Superheavy Elements — the Berkeley Near Miss of 1976

All scientists make mistakes. In fact, it can be said that research is often performed by means of a series of mistakes and corrections. At

Berkeley mistakes were also made, but they were usually never published — until now. Here is the story of how the SHE were “found” for about 18 hours in a great experiment, one wherein the classic tool of nuclear chemistry was used to chemically isolate an element on the shore of the “Magic Island,” or so it was thought. It was a beautiful case of being misled by a subtle artifact and it happened in 1976, within weeks of the report of the ill-fated ORNL/Davis/Florida State experiment. Some of the people primarily involved in this experiment were: LBL — Al Ghiorso, Mike Nitschke, Rollie Otto, David Morissey, Patricia Baisden, Diana Lee, Bob Silva, Malcolm Fowler, and Glenn Seaborg; LLNL — Ken Hulet, Ron Loughheed, John Wild, Jerry Landrum, and Pete Stevenson.

The essence of the experiment was simple, but carrying it out properly was a large task. In 1976, a target of ^{248}Cm (96) was bombarded by ^{48}Ca (20) ions from the SuperHILAC with the hope of making element 116 or 114. Ken and Ron had vaporized a thick target of curium onto a beryllium backing for this bombardment with the hope that it would stand up to the large beam that they expected to use. The beam was not composed of the ordinary atoms of calcium, though, for they had to use the very rare heaviest isotope of that element, ^{48}Ca . This was, and is, a very expensive isotope to use, so it was necessary to make sure that the efficiency of using it in the accelerator was as high as possible. For this purpose Mike and Al spent many hours working with the crew so that this was the case. This meant that they had to adjust the accelerator so that it had the desired energy and they had to make sure that the beam was focussed properly for the experiment. During the week devoted to this particular experiment everything seemed to work out, but it required their full attention as well as that of the crew.

The apparatus in which the bombardment took place was right next to the accelerator to minimize losses, and three charge states of the beam rather than just one were used. This caused more difficulties, since it meant that no magnetic selection was possible and care had to be taken to ensure that all three charge states had the same energy. They also had to be careful that none of the prestripper beam

reached the target, since it would not only have the wrong energy, but would just heat up the target. As for the experiment itself, the reaction recoils were caught in thin aluminum catcher foils adjacent to the target during the long bombardment, and these foils were removed and dissolved. The resulting solution was then subjected to chemical procedures that would give mainly two fractions — a superheavy element fraction and an actinide fraction. A radioactive tracer, ^{241}Am , had been added to the solution so that the overall yield would be known.

Something went wrong with the experiment, however, and a barely visible blackish crud formed that coated the catcher foils. When the foils were dissolved, small amounts of what looked like carbon granules appeared on the surface of the liquid. With great perspicacity Rollie Otto and Mac Fowler realized that if this crud was carbon it could have an important chemical influence on what happened. Since element 116 was expected to have the chemistry of a noble element it, too, would not dissolve and instead would be absorbed by the carbon. To play it safe, they decided that a third fraction should be prepared, a so-called “carbon” fraction, just in case the SHE atoms landed there. To check on this possibility the carbonaceous material was caught on a filter paper as the solution of dissolved activity was passed through it so that it could be analyzed directly with a solid state detector without any additional chemistry.

It was thus that three fractions were separated for analysis. The first, a SHE fraction, according to conventional ideas of the chemistry expected for element 116; the second, an actinide fraction complete with a high yield of the tracer that had been added when the catcher foils were dissolved; and the third, the unplanned one, which could be expected to snare superheavy element atoms if they had the chemistry of noble metals. It was a very clever concept and took advantage of the appearance of the mysterious crud. Now it was up to the analyses of the three fractions.

Not surprisingly, the actinide fraction showed a moderate amount of SF activity; it would contain mostly the well-known ^{256}Fm .

However, in a matter of a few hours it became clear that the "carbon" fraction also had SF activity — indeed, about as much as the actinide fraction had! This was fantastic and extremely exciting news to everyone. And the half-life seemed to be somewhat different, too, but both were in the range of a few hours. It seemed obvious that it could not be an actinide element, since there was no sign of the tracer in the "carbon" fraction. There was no getting around it; they had performed a classical nuclear chemistry separation and placed the SHE atoms in a chemical fraction all by itself by the use of chemistry. There seemed to be absolutely no doubt that this was what had occurred. If their observations were correct they had *chemically* isolated a SuperHeavy Element!

Ghiorso jumped the gun a little by placing a call to Stan Thompson to tell him the exciting news. Al knew that Stan was literally at death's door and thought that he would be pleased to hear that the mission to find the SHE that he had helped to start a dozen years before had finally ended successfully. He was in the last throes of his battle with cancer and could not reply to Al, but Al talked at him and was certain that Stan understood what had been accomplished. He died that evening not very long after Al's call.

Now it was up to the instruments to finish their job. Al remembers having a nagging doubt for some reason and felt only half-convinced. However, he was quite willing to accept the result as a gift of the gods. It was Friday and having spent a strenuous week and with the work done, he went off with his wife, Wilma, to Pt. Reyes, in the afternoon to do some birding, their favorite form of relaxation. He had arranged with Glenn that he would call in later in the day to Berkeley and see how the counting was progressing.

Al reached him about 4:30 p.m. and found that the ratio of the fissions in the "carbon" fraction to those in the actinide fraction was still holding up; now he felt it likely that soon he was going to have to pay off his \$100 bet with Glenn! (Years before, he had made a bet with Glenn that the Superheavies would never be found. It was a bet designed only to be lost. Al would have been more than pleased

now to pay off the bet!) They both felt that the discovery was a very important matter, so they were very cautious and talked with their old wartime code habits by not being very specific about the experiment and using coded words for what they thought had been found. Al remembers that Seaborg was 100% convinced. After all, they had separated a superheavy element chemically, hadn't they?

The following account is given in Al's own words:

"That evening, I went up to the SuperHILAC to check on the counting and make sure that everything was operating properly. I was immediately puzzled by the fact that the two fractions seemed to be decaying at the same rate. About midnight, becoming very suspicious, I arranged it so that I could look at the pulse spectrum of the integral proportional counter that was counting the SFs in the LaF_3 /actinide fraction. I was shocked to find that most of the SFs were not being counted! Apparently, the filter paper, which was very β -active, was producing a 'thick' spectrum that was badly degraded. Because the threshold for making an integral count was set too high, maybe less than 10% of the SFs were being counted. It meant that instead of being comparable, there were 10 or 20 times as many SFs in the actinide fraction as in the carbon fraction. Needless to say, my confidence of SHE in the carbon fraction immediately went down. Something was wrong, but I had no idea what it was. I decided to sleep on it and let my personal computer work on the problem.

"And that it did! Early the next morning I was jolted awake by a truly disturbing *nightmare*. This was the first time in my life that I have had a nightmare caused by an experiment and I hope that it will be the last! It seems that I was driving a car containing other people when I came abreast of another which contained a bunch of Russians. We immediately began racing; and I couldn't pass them on the road for some reason! I was not about to let that stop me, so I drove over onto the shoulder to go around them. In this way I was successful in passing them except that all of a sudden I was confronted by some obstacle (a brick wall, I think). Very much

frightened, I tried to brake the car and found that I couldn't. I realized that we were out of control! At this point I woke up.

"Out of control! In the shower I reviewed the dream and realized the obvious symbolism with our SHE run that was still in progress. I soon came up with a possible explanation for our results. If the carbonaceous films that coated the catcher foils were 5–10% as thick as the aluminum stopper foils, then that same fraction of the recoils would stop in those layers. If that material could not be dissolved, then neither would the actinides lodged inside it. We had not performed any chemistry on these particles at all! I decided that the source of the carbonaceous crud was most likely the organic binder used in the silver paint used to fasten the foils to the rings. I knew that the beam was depositing about 100 watts in the probe during the bombardment so that it would become very hot. It was very likely that the binder would be vaporized and end up as carbon on the aluminum foils. This seemed like a very reasonable scenario so I went up to the Lab to confirm it.

"Shortly after I had arrived and checked the equipment and found that the two fission counters showed the same half-life, Glenn called on the phone to see how the 'SHE' fraction was doing. It must have taken me at least five minutes to convince him that it was a false alarm. Shortly after that Rollie called up, but it took 10–15 minutes to persuade him that the SHE had vanished. I really felt bad for having to deliver the sad news after our having 'discovered' the superheavies for all of 18 hours.

"In retrospect, it was an interesting experience, though very disappointing. We had prepared very well, according to the technology of the times; we had overcome many obstacles just to make the adequate bombardments; we had performed the experiment with professional expertise; we had made an acute observation during the experiment that should have led us to discover the SHE; and, finally, we had chemically separated what could well have been a member of that tribe. Everything was perfect except for that last statement. There was no SHE candidate. We had been misled by an amazing

and also amusing artifact of the experiment. Fortunately, we caught the mistake in interpretation very quickly and nothing was ever published about our 'discovery' until now."

14.7. Future Searches for SHE

It has been proposed recently that the SHE have already been discovered, because in all the elements from fermium on up, the nuclear stability is strongly dependent on the influence of nuclear shells. However, researchers have not yet discovered any new region of elements whose isotopes have lifetimes qualitatively longer than those of their nearest neighbors. In fact, recent calculations have predicted that there is a doubly magic region of extra stability near the deformed nuclear shells at 108 protons and 162 neutrons. The discovery of the longer-lived isotopes of seaborgium of mass 265 (~ 7 s) and 266 (~ 21 s) with 159 and 160 neutrons, respectively, and of the ~ 9 s ^{269}Hs , which has 161 neutrons, tends to confirm this prediction. Should this be called a SHE region? The existence of this new region of deformed stability changes our previous view of the stability of the heaviest nuclei. Spherical nuclei around the hypothetical doubly magic spherical nucleus $^{298}114$ are no longer expected to form an island in a sea of instability. Instead, it appears that there is an extended peninsula of relatively long-lived nuclides leading up to this spherical region. The longest-lived nuclides are now expected to be around the nucleus $^{292}110$ and are predicted to have half-lives of the order of 100 years, long enough to be accumulated, provided ways to produce them can be devised! Many of these SHE are expected to decay predominantly by α -emission, making positive identification easier.

Quoting from a 1977 article^{14,36} by Curt Bemis and Ray Nix entitled "The Quest in Perspective": "As is true with most endeavors worthy of pursuit, the quest for superheavy elements has not been without misfortune. Countless explorers have returned without even a glimpse of the island. Some, perhaps ill-prepared to make the journey, have shouted 'Land ho!' only to have it slowly fade before

their eyes. None of these attempts have ever resulted in conclusive evidence for the existence of superheavy elements. As of this date (1977), there have been a minimum of 15 claims to the discovery of superheavy elements, or at the very least, suggestions that superheavy elements have been responsible for experimental effects. Most, but not all, claims have been retracted or dismissed. Yet the quest must go on!" Unfortunately, we could write the same words today (1998) except that the number of "claims" and negative reports have burgeoned to dozens. However, we have learned much in this pursuit — and in the same spirit and in light of recent theoretical predictions and the development of new, more sensitive techniques, we are not yet ready to give up!

Scientists around the world (Germany, Russia, Japan, US) are considering new searches with more sensitive instrumentation and contemplating what will be the best production reactions to use. In May 1998, researchers at GSI were unsuccessful in their attempt to try to detect element 113 with SHIP in the products of the bombardment of ^{209}Bi with ^{68}Zn projectiles. The Dubna–Livermore collaboration is investigating "hot fusion" reactions in preparation for experiments in which they will try to produce and detect element 114 in bombardments of ^{244}Pu with ^{48}Ca projectiles at the Cyclotron at Dubna. We at Berkeley are currently building a new instrument, the Berkeley Gas-filled Separator (BGS), which should have increased sensitivity for detection of the heaviest elements. The project is led by Ken Gregorich and Victor Ninov and initial testing began in late summer 1998. A workshop was held in May 1997,^{14,37} to discuss the BGS project with potential users and theorists to solicit ideas for planning and implementing a vigorous experimental program. Some of the characteristics of the BGS and the production reactions considered most promising are summarized in Fig. 14.4. It is envisioned that the BGS can also be combined with subsequent chemical separation procedures to study the chemical properties of newly discovered species as well as their nuclear properties. So perhaps within another ten years an epilogue to this chapter can be written on a more positive note!

BERKELEY GAS-FILLED SEPARATOR (BGS)

World-Class Facility at the LBNL 88-Inch Cyclotron

- High-Intensity Beams: Small cross sections accessible
- Large Angular Acceptance: Better efficiency for asymmetric reactions
- Large Bending Angle: Better separation
- Large Momentum Acceptance: High efficiency, thick targets

HEAVY ELEMENT EXPERIMENTS WITH BGS

$^{248}\text{Cm}(^{26}\text{Mg}, 4n)^{270}\text{Hs}$	N=162, Z=108 deformed shells
$^{238}\text{U}(^{36}\text{S}, 4n)^{270}\text{Hs}$	What are α - and SF half-lives?
$^{248}\text{Cm}(^{48}\text{Ca}, 4n)^{292}116$	Spherical Superheavies?
$^{244}\text{Pu}(^{48}\text{Ca}, 4n)^{288}114$	(100 times better sensitivity)
$^{209}\text{Bi}(^{24}\text{Mg}, 3n)^{230}\text{Am}$	Electron-Capture Delayed Fission
$^{209}\text{Bi}(^{28}\text{Si}, 3n)^{234}\text{Bk}$	($Q_{\text{EC}} > \text{Fission Barrier}$)
$^{238}\text{U} + ^{86}\text{Kr}$	Binary Transfers for Production of
$^{248}\text{Cm} + ^{76}\text{Ge}$	New Neutron-Rich Nuclides
	Prior to Chemistry

Fig. 14.4. Characteristics of BGS and some of the most promising reactions for producing new heavy element isotopes and new heavy elements.

Epilogue

It is with the greatest excitement and exhilaration that we add this epilogue to the "Searches for the Superheavy Elements." Our recent successful experiments (April and May 1999) using the BGS at LBNL have provided convincing evidence^{14,38} for the discovery of at least two new superheavy elements, 118 and 116. Thus, at long last, more than 30 years after predictions^{14,3, 14,4} of the Island of Superheavy

Elements in the mid-60s, we have, indeed, reached and even gone beyond the “magic” region around $Z=114$ and $N=184$! When this book was finished, just before Glenn Seaborg left for the Boston ACS meeting, where he suffered his terrible stroke on 24 August 1998, we assumed that all that was left to do was the final proofreading. Then came this fantastic climax to the search for the superheavy elements, a search that had been a prime goal for a large number of scientists, especially for Glenn. We only regret that he is not here to witness it with us.

It began in January 1999 with an alert that a Dubna/LLNL collaboration working at Dubna had observed a single decay chain in the bombardment of ^{244}Pu with ^{48}Ca that they felt could only be attributed to element 114! A first glance at their findings seemed to indicate that, indeed, they had made the long-awaited discovery. Their reported chain of three α -emitters, 30-s, 9.71-MeV $^{289}114$ decaying to 15-min, 8.67-MeV $^{285}112$ decaying to 1.6-min, 8.83-MeV $^{281}110$ decaying to 17-min $^{277}108$ which spontaneously fissioned, was a textbook example of what was expected for a SHE decay. But some doubts about the possibility of random correlations arose, pending observation of another event. The experiment ran for about 40 days until the end of 1998, and with their measured overall yield of about 40%, a cross section of only about a picobarn was indicated! We had anticipated beginning a similar experiment in Berkeley in about eight months. Although we had been scooped in this most important experiment, we felt happy that at last the Magic Island had been found and we redoubled our efforts to get our own experiment under way so that we could confirm the exciting discovery. The BGS had already undergone preliminary testing and a first “real” experiment to measure excitation functions for element 105 isotopes produced in reactions of ^{51}V -projectiles with ^{208}Pb targets had been conducted successfully in December 1998. Now the question was whether to try to repeat the Dubna/LLNL experiment immediately or wait for improvements in the BGS. It soon became obvious that it would not be easy for us to repeat their experiment at once for at

least two reasons: (1) we did not have the multiple ^{244}Pu targets required for the rotating wheel system, nor did we have permission to use these radioactive targets in BGS; (2) we did not have an adequate supply of ^{48}Ca for the required long run, nor was it in the metallic form needed for the cyclotron ion source. This later turned out to have been a serendipitous situation.

At about this time, Robert Smolańczuk brought forward a suggestion that he had communicated to scientists at both GSI and Dubna the year before. Robert, a theoretical physicist who was a student with Adam Sobiczewski at the Soltan Institute for Nuclear Studies in Warsaw, Poland, had earlier published predictions for half-lives of heavy and superheavy elements.^{14,39} He had been at GSI during 1998 and performed semiempirical calculations of production cross sections and had recently published predictions of cross sections for producing superheavy elements.^{14,40} To everyone's surprise, he found that the cross sections did not decrease exponentially with atomic number as had been expected based on extrapolation of the experimental data. In particular, he predicted that the cross section for production of element 118 in bombardment of ^{208}Pb by ^{86}Kr might be as large as 670 pb! This was orders of magnitude larger than previous expectations. During the summer of 1998 he contacted Darleane to inquire whether he could work with the LBNL Heavy Element Nuclear & Radiochemistry Group if he received a Fulbright Visiting Scholar Award. Darleane, who already knew of his research papers, replied that she would be more than happy to have him join the group provided he didn't need any additional support, as there was no money available for another person! She also made it clear that the Group performed experimental research and he replied that he was aware of that, but wanted to work closely with experimentalists. Fortunately for us, Robert Smolańczuk did come to Berkeley as a Fulbright Research Scholar in October 1998. We asked him to acquaint us with his theoretical predictions and he proposed that we try the $^{208}\text{Pb}(^{86}\text{Kr},n)^{293}118$ reaction as soon as possible.

After the Dubna/LLNL report of element 114, several meetings of the low energy group leaders were held at which we debated the pros and cons of trying to repeat the Dubna/LLNL experiment or trying "Robert's reaction" which he predicted^{14,41} would result in the element 118 decay chain shown in Fig. 14.5. Claude Lyneis, in charge of 88-Inch Cyclotron operations, said they could develop and provide the required high intensity beams of 449-MeV ^{86}Kr ions. The ^{208}Pb targets and thin carbon entrance windows could be acquired from GSI; Walter Loveland, on sabbatical leave from Oregon State University, volunteered to supply additional detectors and instrumentation. Darleane and Al strongly urged trying this new reaction as soon as possible rather than trying to repeat the other one with its very low production rate. We might run for months and never see anything, and besides the required targets and projectile materials and Cyclotron time were not available. Ken Gregorich agreed, saying that the BGS efficiency was so high that we could at least push down the limit on the predicted production cross section to the few picobarns level even if we were not successful in detect-

$^{208}\text{Pb}(^{86}\text{Kr}, 1n)^{293}118 \quad \sigma = 670 \text{ pb}$		
$E_{\text{LAB}} = 449.0 \text{ MeV} \quad E_{\text{CM}} = 317.7 \text{ MeV} \quad E' = 13.3 \text{ MeV}$		
^A_ZN	$Q_{\alpha} \text{ (MeV)}$	T_{α}
$^{293}118_{175}$	12.23	31 μs - 310 μs
$^{289}116_{173}$	11.37	960 μs - 9.6 ms
$^{285}114_{171}$	11.18	800 μs - 8.0 ms
$^{281}112_{169}$	11.00	610 μs - 6.1 ms
$^{277}110_{167}$	10.77	620 μs - 6.2 ms
$^{273}\text{Hs}_{165}$	9.69	120 ms - 1.2 s
$^{269}\text{Sg}_{163}$	8.35	8.0 - 80 min
$^{265}\text{Rf}_{161}$		$T_{\text{st}} \sim 41 \text{ min}$

Fig. 14.5. Decay sequence for $^{293}118$ predicted by R. Smolańczuk.^{14,41}

ing element 118 and its decay chain. Furthermore, it would give us some useful experience with the new system. We finally all agreed to "go for it" and convinced Victor Ninov that the reaction should be run as soon as humanly possible as we greatly feared GSI or Dubna might do it first!

The experiment started on April 8, 1999, and ran for about four days with lots of things going wrong, as might be expected. It was a continuing fight for Ninov, Gregorich, and Loveland to find and fix problems. A Parallel Plate Avalanche Counter (PPAC) was added to discriminate between heavy particles coming through the BGS and α -decays in the focal plane detector. In addition, a second silicon-strip "punch-through" detector was placed behind the focal plane detector to reject events from high-energy light particles coming through the BGS. It was a good "dry run" for a while. But then there was an intriguing event, a unique series of α -particles with energies of 10–12 MeV that occurred in less than a second, something that had not been seen in any previous runs! Could it be that this was the chain of α -decays that had been predicted? And then later it occurred again! Now Victor devoted himself to analyzing the data ever more carefully to make sure that there was not some obscure glitch. He soon satisfied himself that the data from the experiment fit the predictions of Smolańczuk and that elements 118, 116, and 114 had been found! On Monday morning, April 19, Darleane received a phone message from Ken Gregorich asking if he, Victor, and Walter could come up to her office right away and meet with her. She envisioned that something horrible must have happened, but Ken reassured her that was not the case. Victor had been going over the data from the April experiment and had found three possible element 118 decay chain candidates! Two of these were particularly convincing, showing unique high-energy α -decay sequences with lifetimes very close to those predicted by Robert, causing Victor to inquire, "Does Robert talk to God or what??" Anyway, we decided to keep this information quiet until a second experiment could be scheduled and performed to confirm these results — also, some

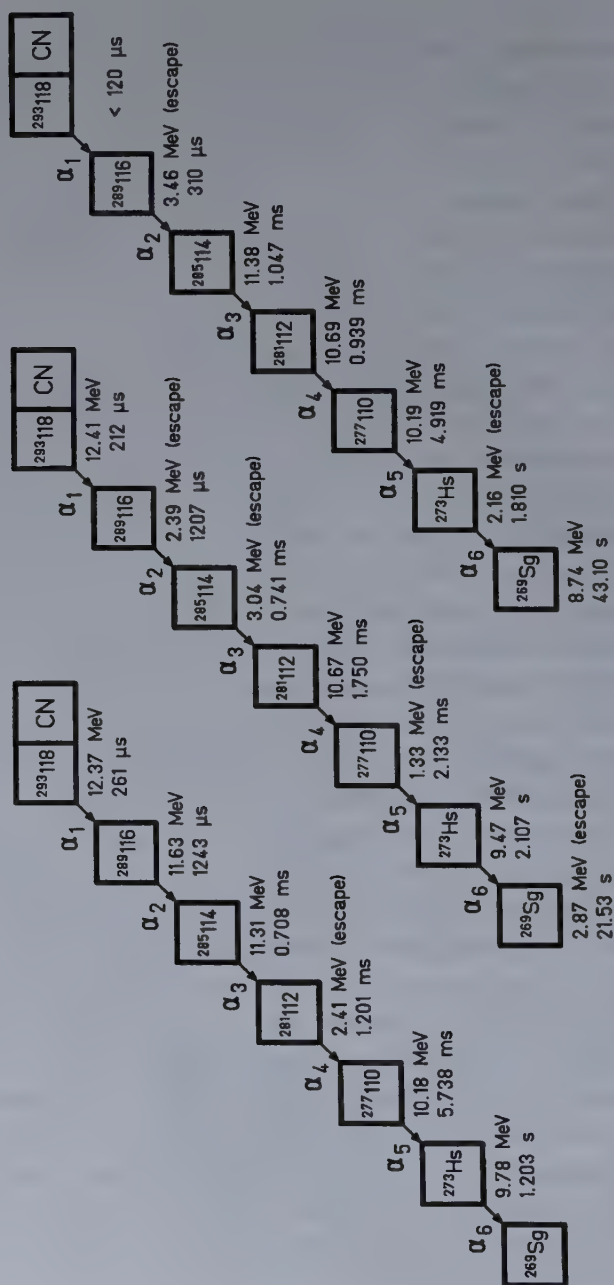


Fig. 14.6. Three decay chains observed by Ninov *et al.*^{14,38} for reaction of 449-MeV ^{86}Kr with ^{208}Pb .

improvements to the BGS detection system would be made. A few people at the Cyclotron and our Division Leader Lee Schroeder and Director Chuck Shank were informed of the potential discovery.

A short time later, on April 30, the second experiment began after the addition of a "punch-through" detector that subtended the entire focal plane and some other minor improvements. This time only one chain was observed over the next six days even though the integrated beam was larger than in the first experiment. Two possible reasons for this are: (1) the Cyclotron energy was not quite the same; (2) it was just a statistical variation. The important thing was that the new chain was an excellent one and confirmed the previous ones. The cross section was estimated to be about 2 pb. The three decay chains observed in the reaction of 449-MeV ^{86}Kr with ^{208}Pb are shown schematically in Fig. 14.6. Based on these data, the half-lives of the chain members were estimated to be 0.12 ms, 0.60 ms, 0.58 ms, 0.89 ms, 3.0 ms, and 1.2 s for elements 118, 116, 114, 112, 110, and 108, respectively. It was such a startling discovery that strenuous efforts were made to find out if anything had gone wrong, but nothing obvious was uncovered. Needless to say, the news was an enormous surprise to the scientific world. Now there is no question, the SuperHeavy Island actually exists! Thus, even if the Dubna/LLNL report of element 114 is not substantiated, we have convincing evidence for elements 114, 116, and 118!! This opens up a whole new region for study. Of course, the obvious next step is to use ^{209}Bi targets to produce the element 119–117–115–113 decay chain, giving us four more new superheavies, although the production rate will probably be smaller. According to Robert, an even better possibility might be to use ^{87}Rb projectiles with ^{208}Pb targets. Thus we close this Epilogue on a note of great optimism and high expectations for the future.

Darleane Hoffman

Albert Ghiorso

13 June 1999

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Chapter 15

Reflections and Predictions

There are almost unlimited possibilities for future research on transuranium elements, which already have extended by more than 20% the total of known chemical elements. The time-line in Fig. 1.5 shows the year when each of these was discovered. Truly, we have been in a golden age for the discovery of new chemical elements! When thinking in terms of the distant future, the tendency is to underestimate the potential for future contributions.

Of central importance is a national policy discussion to retain and recover the potential stock of heavy transuranium nuclides now in storage basins at the Savannah River Plant in South Carolina. These are a priceless treasure and should not be discarded as irretrievable waste. This store, which includes some 7 kg of ^{243}Am and 2.5 kg of ^{244}Cm , can serve as starting material for the synthesis of increasing amounts of heavier transuranium nuclides for years to come.

Estimates suggest that 500 transuranium nuclides would have half-lives sufficiently long to be detectable experimentally (longer than a μs). The synthesis and identification of another half-dozen or so elements seem likely; this would include the discovery of SHE and the extension of the present peninsula of elements to connect with the "island of stability" which would be centered on element 114. Longer-lived isotopes than those now known will probably be found in the transactinide region, especially among the early transactinide elements. The recently discovered longer-lived isotopes of lawrencium (element 103) (^{261}Lr and ^{262}Lr , with half-lives of 39 and 216 minutes, respectively) will make possible a detailed study of the chemical properties of this element. It should be possible to

study the chemical properties of elements beyond seaborgium (element 106).

Figure 15.1 shows an imaginative periodic table extending all the way up to element 168. The completion of the actinide series, with the filling of the 5f electron subshell, occurs at element 103 (lawrencium). Elements 104 (rutherfordium) through 112 are formed by filling the 6d electron subshell, which makes them homologs in chemical properties with the elements hafnium ($Z=72$) through mercury ($Z=80$). Elements 113 through 118 would result from filling of the 7p subshell and are expected to be similar to the elements thallium ($Z=81$) through radon ($Z=86$). The 8s subshell should fill at

1 H																	2 He				
3 Li	4 Be															5 B	6 C	7 N	8 O	9 F	10 Ne
11 Na	12 Mg															13 Al	14 Si	15 P	16 S	17 Cl	18 Ar
19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr				
37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe				
55 Cs	56 Ba	57 La	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn				
87 Fr	88 Ra	89 Ac	104 Rf	105 Ha	106 Sg	107 Bh	108 Hs	109 Mt	110	111	112	(113)	(114)	(115)	(116)	(117)	(118)				
(119)	(120)	(121)	(154)	(155)	(156)	(157)	(158)	(159)	(160)	(161)	(162)	(163)	(164)	(165)	(166)	(167)	(168)				

LANTHANIDES

58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb	71 Lu
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ACTINIDES

90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No	103 Lr
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SUPER-
ACTINIDES

(122)	(123)	(124)	(125)	(126)											(153)
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Fig. 15.1. Futuristic periodic table of the elements projected out to element 168, with the atomic numbers of undiscovered elements in parentheses. Included are the undiscovered elements 122 through 153, which would constitute the predicted and probably unattainable "superactinide" series.

elements 119 and 120, thus making these an alkali and alkaline earth metal, respectively. Next should come the filling of the 5g and 6f subshells, 32 places in all, which are termed the "superactinide" elements, followed by the filling of the 7d subshell (elements 154 through 162) and 8p subshell (elements 163 through 168).

Although we can feel confident that this is the approximate form the periodic table should assume, we, unfortunately, will not be able to verify much of this experimentally, because the half-lives of most of the nuclei are too short and there are no nuclear synthesis reactions available to reach such heavy elements. However, we believe, as indicated, that it will be possible to add (at least) some six new known elements (perhaps slightly more) to our periodic table.

Much more research on the macroscopic properties of einsteinium (element 99) will be possible with the availability of ^{254}Es (half-life of 276 days) in microgram amounts. It will surely be possible to study the macroscopic properties of fermium (element 100) and not out of the question that this will eventually be done for mendelevium (element 101). The art of one-atom-at-a-time chemistry will advance far beyond what can be imagined today to make it possible to study the chemistry of heavier and heavier elements. All of this will result in the delineation of relativistic effects on the chemical properties of these very heavy elements, which might thus be substantially different from those expected by simple extrapolation from their lighter homologs in the periodic table.

Such a research program will require, for success, the availability of apparatus and equipment of increasing complexity, versatility, and power. Essential will be the need for higher neutron flux reactors for sustained operation as research tools and to produce large quantities of transplutonium nuclides for use in research on their properties and as target materials for production of the presently known (see Fig. 15.2) and predicted new heavy element nuclides. Higher neutron fluxes will be especially valuable for the production of the heaviest nuclides, ^{254}Es and ^{257}Fm , which can provide springboards to the region beyond. In fact, in 1984 a consortium of LBL, ORNL, LANL, and LLNL scientists submitted a proposal^{15.1} to DOE

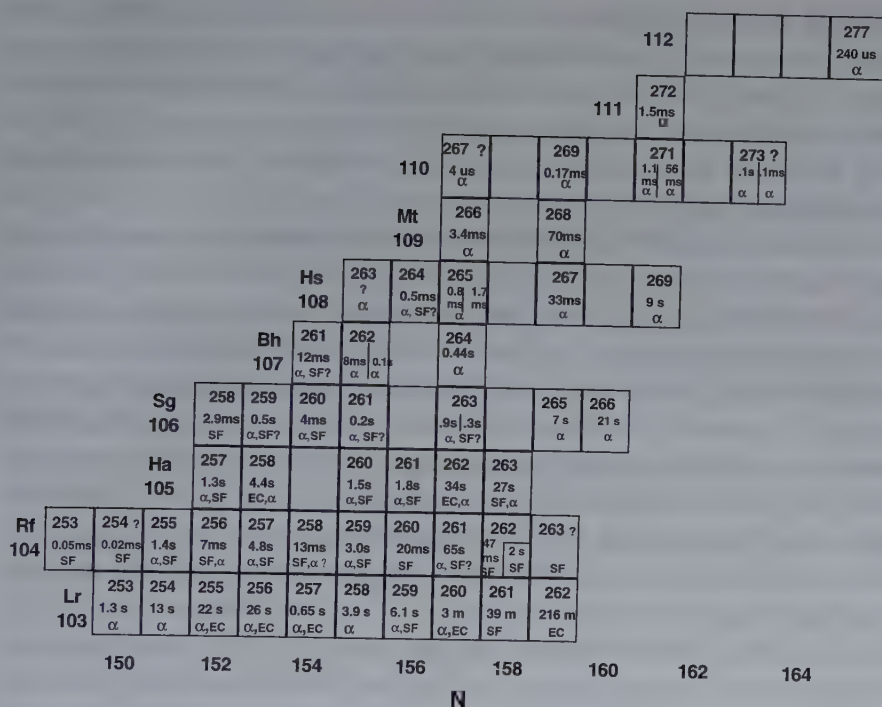


Fig. 15.2. Chart of known transnobelium isotopes as of mid-1997.

entitled "The Large Einsteinium Accelerator Program (LEAP)" — "to investigate the inorganic and nuclear chemistry of elements in the transfermium region, including a search for SHE." Central to this program were two campaigns at the HFIR (planned two years apart) to produce at least 40 μg of 276-d ^{254}Es . The three main goals were: "(1) a new search for SHE by bombarding targets of the ^{254}Es with ^{48}Ca , (2) a major effort to determine the chemical properties of the short-lived elements in the transeinsteinium region, and (3) a comprehensive search for and characterization of new neutron-rich isotopes in this region." A follow-on campaign to produce 40-d ^{255}Es was also outlined which would take advantage of the still higher cross sections for production of neutron-rich isotopes. Some initial

funding for the program was obtained for exploratory experiments, but, unfortunately, full funding was not forthcoming, and production and use of the proposed large sources of einsteinium was never realized. However, this effort did spark a renewed interest in achieving the goals of this project using the available lighter radioactive targets, such as $320\text{-d }^{249}\text{Bk}$.

Higher intensity medium-to-heavy ion accelerators must be built and the means of coping with the heat generated in the target by such intense beams must be developed in order to overcome limitations due to small nuclear reaction cross sections. Increases by orders of magnitude in heavy ion intensity should make possible nuclear synthesis reactions with secondary (radioactive) beams of neutron excess projectiles, which might greatly increase the yields of sought-after new nuclides. Improved methods for safe and efficient handling techniques for making chemical measurements on increasingly larger quantities of the highly radioactive transcurium nuclides must also be developed.

Currently, there are new developments in the chemistry and physics of the heaviest elements, both in theory and in experiment. Those of us who have been active over the past ten years in the new field of applying one-atom-at-a-time chemistry to the exploration of the chemical properties of the heaviest elements ($Z > 101$) are extremely excited about the recent experimental evidence which seems to confirm the prediction of enhanced nuclear stability due to the deformed nuclear shells at $Z=108$ (hassium) and $N=162$! ^{270}Hs has been predicted to α -decay with a half-life of about 6 s. If current theoretical predictions are correct, a whole new vista of longer-lived isotopes will become available for the study of both chemical and nuclear properties of isotopes of elements perhaps up to element 111, assuming we can devise appropriate reactions and increase our overall yields, perhaps by the use of multiple target arrangements. This optimistic outlook is in contrast to the situation some years ago, when it was believed that SF half-lives would become so short (until we reached the spherical doubly magic region around $Z=114$ and

N=184) that it would be nearly impossible to observe the α -decay or study the chemical properties of elements beyond 106.

These are especially exciting times for both the chemistry and physics of the transactinides. New separator systems such as the Berkeley Gas-filled Separator being built under the direction of Ken Gregorich and Victor Ninov should help us determine the half-lives and nuclear properties of predicted longer-lived isotopes of the elements with $Z > 106$ prior to beginning the chemical studies. They will also facilitate searches for elements with $Z > 112$ and permit us to investigate the best reactions for reaching the region of the doubly magic spherical nucleus $^{298}114$ which Smolanczuk^{15.2} has predicted will α -decay with a half-life of about 10 m (SF half-life $> 10^5$ years). He also predicted that $^{292}110$ will α -decay with a half-life of about 50 years (SF half-life $> 10^5$ years), so it could be built up in long irradiations and then separated and measured. Another important experiment will be to confirm the existence of the doubly magic deformed nucleus ^{270}Hs .

Relativistic effects increase proportionally with Z^2 , and it now appears that a whole new region for chemical research is before us which will allow us to explore more fully the range of relativistic effects on chemical properties which we have only begun to glimpse so far. Furthermore, as is the case with the nuclear studies, we can now compare the experimental results with the newly developing relativistic theoretical calculations. The proceedings^{15.3} of a recent conference on "The Transactinide Elements," sponsored by the Welch Foundation, give an excellent picture of the current state of knowledge concerning both the chemical and nuclear properties of these elements and capture the excitement and optimism concerning the prospects for producing longer-lived isotopes of the known elements, and for discovering elements beyond the currently known 112.

In January 1991, on the anniversary of the discovery of plutonium, Roger Batzel, former Director of LLNL, speaking for LLNL and LBL in collaboration with the University of California/Berkeley, announced the establishment of the Glenn T. Seaborg Institute for

Transactinium Science. The Institute Charter was approved by David P. Gardner, then President of the University of California, and signed by Director John H. Nuckolls, LLNL, and Director Charles V. Shank, LBL. The Institute is devoted to the study of the transactinium elements, with special emphasis on the education and training of future generations of scientists in heavy element research. It has provided a unique focus and mechanism for cooperation and collaboration between the system-wide campuses and laboratories of the University of California and with the transactinium community worldwide. Darleane Hoffman was appointed to be the first Director and was succeeded by Louis J. Terminello in late 1996. Another branch of the Seaborg Institute was established in 1997, at the Los Alamos National Laboratory, with David Clark as Director. It is our dream that in the future additional laboratories or institutes will be created for exclusive research on the transactinium elements.

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